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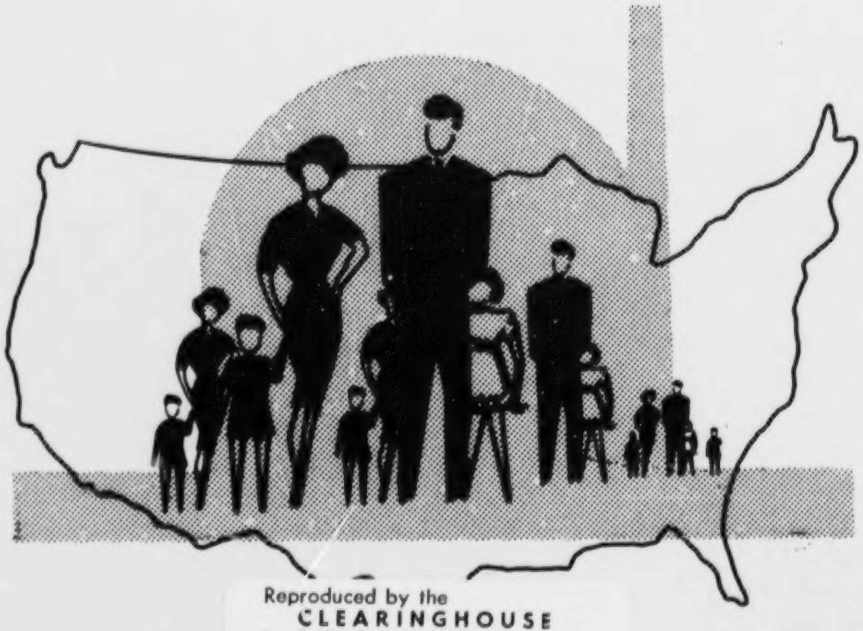
RADIOLOGICAL SURVEILLANCE STUDIES AT A BOILING WATER NUCLEAR POWER REACTOR



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U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
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RADIOLOGICAL SURVEILLANCE STUDIES AT A BOILING WATER NUCLEAR POWER REACTOR

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U. S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
Public Health Service
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FOREWORD

The Bureau of Radiological Health carries out a national program designed to reduce the exposure of man to hazardous ionizing and non-ionizing radiation.

Within the Bureau, the Division of Environmental Radiation conducts programs relating to 1) public health evaluation of planned and operating nuclear facilities, 2) field studies at operating nuclear facilities to develop environmental surveillance technology, and 3) systems of national radiation surveillance programs to evaluate population exposure from all sources of environmental radioactivity.

The Bureau publishes its findings in *Radiological Health Data and Reports* (a monthly publication), Public Health Service numbered reports, appropriate scientific journals, and Division technical reports.

The technical reports of the Division of Environmental Radiation allow comprehensive and rapid publishing of the results of intramural and contractor projects. The reports are distributed to State and local radiological health program personnel, Bureau technical staff, Bureau advisory committee members, university personnel, libraries and information services, industry, hospitals, laboratories, schools, the press, and other interested individuals. These reports are also included in the collections of the Library of Congress and the Clearinghouse for Federal Scientific and Technical Information.

I encourage the readers of these reports to inform the Bureau of any omissions or errors. Your additional comments or requests for further information are also solicited.

John C Villforth

John C. Villforth
Bureau Director

PREFACE

The projected increase in the utilization of nuclear power for electrical generating plants has resulted in both State and Federal public health agencies placing increased program emphasis on the surveillance of nuclear power plants. The Bureau of Radiological Health provides recommended nuclear facility surveillance program information for the guidance of health agencies. In order to provide a better technical basis for our surveillance recommendations, a series of field studies were conducted at operating nuclear facilities to obtain better data on radionuclides in plant effluents and their subsequent distribution in the environment.

This technical report summarizes the first such study which was conducted at the Dresden Nuclear Power Station. The study was planned and performed by the staff of the Division of Environmental Radiation, Bureau of Radiological Health, Environmental Control Administration, with close cooperation of the Illinois Department of Public Health, the Commonwealth Edison Company, the Division of Compliance, AEC, and the Division of Radiation Protection Standards, AEC.

We wish to thank all of the reviewers of this report for their extensive comments on the preliminary draft.

Charles L. Weaver

Charles L. Weaver
Director, Division of Environmental
Radiation

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1. INTRODUCTION

1.1 Need for Study

Radiological surveillance at central-station electric power reactors under normal operating conditions provides assurance of adequate control over radionuclide effluents and a means for estimating the resulting radiation exposure of the population. The purpose of this study was to identify and quantify the radionuclides in effluents and in the pathways from the point of discharge in the environment to man in order to provide the technical basis for improving surveillance programs. Improvements are needed because of the projected increase of commercial nuclear power stations in the United States.(1) The current interest of the public in reducing environmental pollution and of the industry in conducting more informative monitoring programs lends support to the aims of this study.

Monitoring fallout from the nuclear weapon tests and discharges at atomic energy research laboratories has provided extensive knowledge and experience. These are reflected in the many publications on environmental monitoring,(2-23) radioactive waste management,(24-30) and radionuclide transfer in the biosphere.(31-45) Some information on waste management is also available in the periodic reports by commercial operators of nuclear power plants to the Atomic Energy Commission (AEC).

The Bureau of Radiological Health has begun a series of studies at central-station electric power reactors with the following specific objectives: (1) to gain information on individual radionuclides in the effluent as a basis for developing recommended radiological surveillance programs, (2) to evaluate programs associated with measuring discharged radionuclides and interpreting results in terms of radiation exposure, (3) to observe the movement of critical radionuclides from a plant through the environment under routine conditions of station operation, (4) to develop competence within the Bureau of Radiological Health for assisting public agencies in monitoring sites of nuclear power plants. It should be noted that the studies are not intended to investigate waste management or radiological surveillance practices at specific nuclear power stations.

1.2 Study Design

The first radiological surveillance study in this program, described here, was undertaken at the Dresden Nuclear Power Station of the Commonwealth Edison Company. Dresden I is a boiling water reactor (BWR) that has generated more than 9 x 10^9 kilowatt-hours since it began operation in 1959, and has been operating at a rated power of 700 thermal megawatts (MWt) and 210 megawatts of electricity (MWe) since 1962. The power plant is a dual-cycle, forced circulation system, built by the General Electric Company. Fuel elements consist of slightly enriched UO_2 clad in Zircaloy-2. The station is located in Illinois, 80 km SW of Chicago. Liquid wastes are discharged into the cooling-water discharge canal which empties into the Illinois River; gases and airborne particles are released from a 91-m stack; and solid wastes are transported off-site for burial.

The study was planned and performed by the staff of the Division of Environmental Radiation, Bureau of Radiological Health, Environmental Health Service, PHS, with the close cooperation of the Illinois Department of Public Health, Commonwealth Edison Company, Division of Compliance of the AEC, and Division of Radiation Protection Standards of the AEC. The data were collected in five field trips on November 14–16, 1967, January 16–18, 1968, January 31–February 1, 1968, June 25–28, 1968, and August 20–22, 1968. Participants in the field trips are listed in Appendix A. Results were reported to all participants soon after the field trips and have been reviewed by them.

Emphasis was placed on relating discharges to environmental levels of radionuclides and radiation, and on evaluating critical pathways and radionuclides. The greatest efforts were devoted to measuring the concentration of individual radionuclides in liquids and gases on site and in effluent liquids, gases and airborne particles; determining the radiation exposure rate and radionuclide concentration in air at ground level beneath the plume from the stack; and testing devices that concentrate radionuclides in air and water for subsequent analysis. Other environmental samples were analyzed, but further work is needed in this area to provide more specific information and to attain greater analytical sensitivity. The study was planned to avoid duplicating the following active monitoring programs: (1) gross activity in the effluents measured by Dresden staff, (2) environmental radionuclides measured by Dresden's surveillance contractor, (3) environmental gross activity measured by the Illinois Department of Public Health, and (4) the Aerial Radiological Measuring Survey conducted by an AEC contractor.

Similar studies are in progress at a pressurized water reactor and limited studies are planned for some of the larger stations that are now being placed into operation. These studies will be directed toward evaluating differences in the radionuclide content of the liquid and gaseous effluents and the transfer through the biosphere to man.

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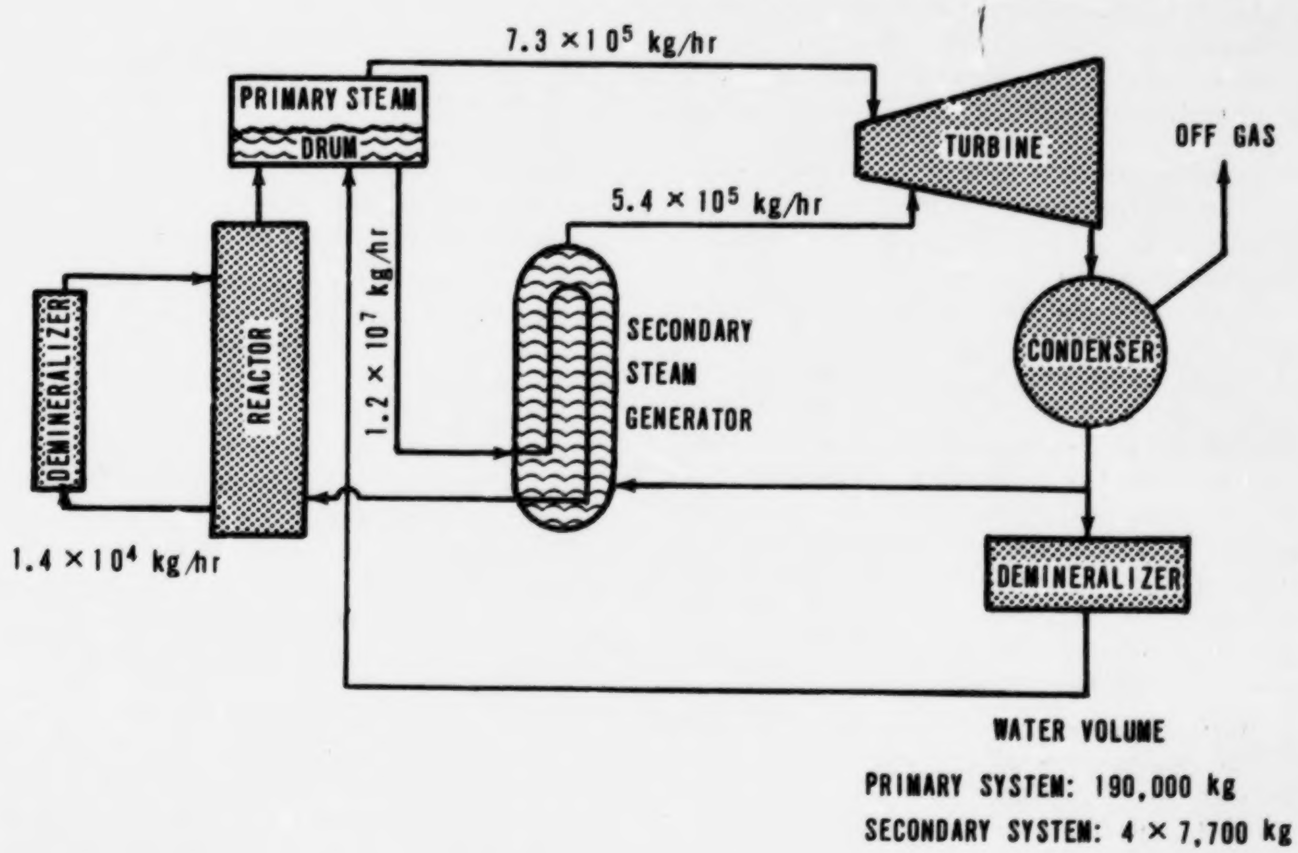
2. RADIONUCLIDES IN LIQUIDS ON SITE

2.1 Samples

2.1.1 *Collection.* Radionuclides were measured in primary coolant, fuel pool, and contaminated demineralized water to identify them in a relatively concentrated state, and to compare concentrations on site and in effluents. The first sample of primary coolant water was obtained at the water sampling port of the primary steam drum on February 1, 1968. On August 22, 1968, two more samples of primary coolant water (one acidified to 0.6 N in HCl to reduce radiocolloidal losses), one sample of fuel pool water, and one of contaminated demineralized water were obtained. Dresden staff provided 1-liter volumes of all samples in polyethylene bottles.

2.1.2 *Primary coolant.* This sample is representative of 1.9×10^5 liters (50,000 gal) of low conductivity demineralized water that circulates as shown in the simplified schematic drawing of Figure 2.1. In brief, steam is generated in the reactor and the secondary steam generators, produces mechanical energy for generating electricity in passing through the turbines, and is then condensed for recirculation. Material



7.3 × 10⁵ kg/hr
PRIMARY STEAM
DRUM
TURBINE
OFF GAS
5.4 × 10⁵ kg/hr
1.2 × 10⁷ kg/hr
DEMINERALIZER
REACTOR
SECONDARY
STEAM
GENERATOR
CONDENSER
1.4 × 10⁴ kg/hr
DEMINERALIZER
WATER VOLUME
PRIMARY SYSTEM: 190,000 kg
SECONDARY SYSTEM: 4 × 7,700 kg

Figure 2.1. Primary coolant flow schematic.

may leave the system during shut-down when it is opened for refueling or repair, and during routine operation by the following paths:

1. Gas flows from condenser air ejectors through a delay line and two filters in series to the stack for release; minor fractions of gas reach the stack from gland-seal condenser air ejectors through a delay line, and from leaks to the ventilating air.
2. Water is removed in the course of blowdowns, escapes at leaks as steam or water, and is carried with released gas; it is recovered and reused except for high-conductivity liquid waste and water vapor in the stack effluent.
3. Ionic and solid impurities are removed in the course of periodic replacement of the reactor water demineralizers (the used resins are shipped for burial as solid waste); weekly regeneration of condenser-water demineralizers (the regenerating solution is discharged as high-conductivity liquid waste); periodic replacement of filters and low-conductivity-water demineralizers; and blowdown of each of four secondary steam generators (the blowdown water is filtered, demineralized, and reused).

Fission products enter the primary coolant water from fuel elements through holes or cracks in the cladding, or from uranium that was released from failed fuel elements. Tramp uranium and diffusion through intact cladding are believed to be minor sources of fission products.(1) At Dresden, much of the fission product activity in primary coolant water is attributed to uranium that had entered the primary coolant several years previously from failed fuel elements.(2) Activation products are formed in the reactor by irradiation—mostly with neutrons—of air, coolant water, and corrosion products, erosion products, and water impurities in the coolant.

Radionuclides are removed from the water in the reactor vessel by radioactive decay, deposition on surfaces and accumulation as "crud" in recesses,(3) ion exchange and filtration on the reactor-water demineralizer, and carryover with steam to the turbines and condensers. At the condenser, most non-gaseous radionuclides, including the particulate decay products of the radioactive gases that are formed as the gases are carried from reactor to condensers, remain in the condensate.(1) Accumulation occurs in the condenser-water demineralizer and secondary-steam-generator water. Most particulate radionuclides that are carried with or formed from the gases decay within the delay line, and others are flushed from the delay line with condensed moisture;(1) those that pass through the delay line are removed on the filters to the extent of filter efficiencies before entering the stack. Radioactive gases are discharged in gaseous effluent at the top of the stack, together with particulate radionuclides that (a) are descendants of these gases and have been formed after filtration, (b) penetrated the filters, and (c) are carried in unfiltered gas streams (i.e., ventilating air and gland-seal condenser off-gas).

2.1.3 *Fuel storage pool.*(2) The pool contains 3.8 x 10^5 liters (100,000 gal) of demineralized water, and is an independent system during reactor operation. The water is circulated through a filter to remove suspended material and insoluble radionuclides. Longer-lived fission products leached from the stored fuel elements either circulate in the water, are filtered, or deposit in the fuel pool system.

When the reactor is being refueled, the refueling canal is flooded with demineralized water to combine the reactor vessel, the refueling canal, and the fuel pool into a single body of water. Water from the bottom of the reactor vessel is continuously blown down to the radioactive liquid waste treatment system during this operation. This water is filtered, demineralized, and returned to the refueling canal. The gross radioactivity concentration in fuel pool water is maintained below 1 x 10^{-4} μC/ml by Dresden.

2.1.4 *Contaminated demineralized water.*(2) Approximately 7.6 x 10^5 liters (200,000 gal) of water that has been reprocessed is stored for reuse. This water enters the storage tank by being rejected from the demineralized condensate water when the water level rises in the condenser hot well, or by transfer of filtered and demineralized liquid radioactive waste.

2.2 Analysis

2.2.1 *Gamma-ray spectrometry.* Radionuclides that emit gamma rays were identified and quantified in sample aliquots by multichannel spectrometry with a Ge(Li) detector, as shown in Figures 2.2 and 2.3. Decay measurements with eight spectra obtained from 0.5 to 150 days after sample collection were used to measure long-lived radionuclides without interference by short-lived ones and to confirm identification by gamma-ray energies. The minimum half life of radionuclides detected by this procedure was 6 hours, and the maximum, 30 years. Minimum detectable concentrations at these half lives were approximately 5×10^{-4} and 1×10^{-5} microcurie per milliliter (μCi/ml), respectively.

2.2.2 *Alpha-particle spectrometry.* Radionuclides that emit alpha particles were analyzed in evaporated aliquots by spectrometry with a Si-diode detector, and quantified with a proportional counter. Only a single alpha-particle group at 6.11 ± 0.02 MeV could be definitely identified in the primary coolant; this was attributed to ^{242}Cm.

2.2.3 *Radiochemistry.* All of the expected radioelements except rhodium and curium were separated chemically and then measured with appropriate radiation detectors. Radiochemical analysis confirmed spectral identification by gamma-ray energy and half life, was more sensitive than instrumental analysis by itself, and provided values for radionuclides that emit no gamma rays, or whose gamma rays were obscured in the spectra. Thus, ^{89}Sr and ^{90}Sr were measured with a low-background G-M counter, tritium, with a liquid scintillation counter after distillation, and ^{55}Fe, with a xenon-filled proportional counter plus spectrometer. Radionuclides that emit gamma rays were measured after chemical separation with a NaI(Tl) detector plus spectrometer. Aliquots of 10–20 ml were usually analyzed. Because of improved techniques, results for the primary coolant sample of August 22 are more precise than for the earlier sample.

2.3 Results and Discussion

2.3.1 *Radionuclides in primary coolant.* Most high-yield fission products with half lives of 6 hours or longer were identified and measured, as well as activation products with high (n,γ), and (n,p) cross-sections in water, steel, Zircaloy, copper, and silver (see Table 2.1). The short-lived radionuclides at highest concentration were ^{135}I and ^{99m}Tc; the longer-lived ($t_{1/2}$ >1 week) radionuclides at highest concentration were ^{131}I and ^{58}Co. The radionuclides ^{59}Fe, ^{82}Br, ^{115m}Cd, and ^{127}Sb were not detected at levels of 1×10^{-6} μCi/ml, and ^{32}P was less than 1×10^{-5} μCi/ml.

The sum of measured radionuclide concentrations was 0.4 μCi/ml on February 1 as compared to 0.13 μCi/ml on August 22, and most individual radionuclides in the later sample were at lower concentrations. The lower concentrations could have resulted from the shorter operating period since the preceding partial refueling (see Figure 2.4), from better-clad fuel elements added at the fifth partial refueling, or from cleanup of the primary coolant system in the interval. The activation product analyses confirm earlier reports for Dresden,(3,4,5,6) and for a similar reactor at Garigliano,(7) except that measured concentrations are somewhat higher. The earlier reports also gave values for the short-lived radionuclides 1.8-hr ^{18}F, 2.6-hr ^{65}Ni, 2.6-hr ^{56}Mn, and 0.9-hr ^{134}I; these were not found in this study because of the relatively long time interval between sampling and analysis. Tritium was not reported in the earlier studies. Tritium concentrations of 7×10^{-3} and 3×10^{-3} μCi/ml in 1965(5,8) were somewhat higher than given here. The presence of ^{242}Cm had not been reported, but it was predicted as the alpha-emitting transuranium radionuclide with the highest decay rate in fuel after reactor operation for 1 to 2 years.(9)

2.3.2 *Radionuclide losses in samples.* The walls of the polyethylene bottles retained detectable amounts of the radionuclides from unacidified primary coolant

Table 2.1

Radionuclide Concentration in Primary Coolant*

Radionuclide	Concentration, μCi/ml	
	Feb. 1, 1968	Aug. 22, 1968
from fuel		
51 -d ^{89}Sr	1.2×10^{-4}	4.4×10^{-5}
28 -yr ^{90}Sr	1.2×10^{-5}	1.3×10^{-6}
9.7 -hr ^{91}Sr	2.4×10^{-2}	1.2×10^{-2}
10.3 -hr ^{93}Y	$\sim 7 \times 10^{-3}$	$\sim 2 \times 10^{-3}$
65 -d ^{95}Zr**	9×10^{-4}	2.4×10^{-5}
17 -hr ^{97}Zr**	1.0×10^{-2}	1.3×10^{-4}
35 -d ^{95}Nb**	5×10^{-4}	1.8×10^{-5}
66.3 -hr ^{99}Mo**	1.4×10^{-3}	1.4×10^{-3}
6.0 -hr ^{99m}Tc**	9.0×10^{-2}	4.4×10^{-2}
39.7 -d ^{103}Ru	5×10^{-4}	5.6×10^{-5}
1.0 -yr ^{106}Ru	NM	1.3×10^{-6}
36 -hr ^{105}Rh	$\sim 4 \times 10^{-3}$	$\sim 4 \times 10^{-4}$
78 -hr ^{132}Te	8×10^{-4}	1.9×10^{-5}
8.06-d ^{131}I	7.8×10^{-3}	2.2×10^{-3}
20.9 -hr ^{133}I	6.6×10^{-2}	2.3×10^{-2}
6.7 -hr ^{135}I	1.3×10^{-1}	3.6×10^{-2}
2.07-yr ^{134}Cs†	1.3×10^{-5}	2.3×10^{-5}
13 -d ^{136}Cs†	3×10^{-5}	2.4×10^{-5}
30 -yr ^{137}Cs	3×10^{-5}	4.4×10^{-5}
12.8 -d ^{140}Ba	1.4×10^{-3}	1.0×10^{-3}
32.5 -d ^{141}Ce	9×10^{-4}	4.4×10^{-5}
33 -hr ^{143}Ce	3.2×10^{-3}	1.0×10^{-4}
284 -d ^{144}Ce	3×10^{-4}	8.0×10^{-6}
11 -d ^{147}Nd	3×10^{-4}	$\sim 3 \times 10^{-5}$
2.34-d ^{239}Np†	2.1×10^{-2}	4.1×10^{-3}
163 -d ^{242}Cm†	2.2×10^{-7}	1.2×10^{-7}
from water, cladding, and construction materials		
12.3 -yr ^{3}H‡	1.7×10^{-3}	1.3×10^{-3}
15.0 -hr ^{24}Na	3.0×10^{-3}	1.6×10^{-3}
27.8 -d ^{51}Cr	NM	$\sim 5 \times 10^{-4}$
313 -d ^{54}Mn	NM	1.6×10^{-6}
2.7 -yr ^{55}Fe	9.5×10^{-5}	4.0×10^{-5}
270 -d ^{57}Co	$\sim 1 \times 10^{-5}$	1.6×10^{-6}
71 -d ^{58}Co	1.4×10^{-2}	1.7×10^{-3}
5.26-yr ^{60}Co	2.2×10^{-3}	2.6×10^{-4}
12.7 -hr ^{64}Cu	$\sim 1 \times 10^{-2}$	2.2×10^{-3}
244 -d ^{65}Zn	NM	4.0×10^{-6}
253 -d ^{110m}Ag	NM	1.9×10^{-6}
115 -d ^{182}Ta	2×10^{-5}	$\sim 7 \times 10^{-7}$

*Concentrations at time of sampling.

**Also activation products.

†Formed by (n,γ) reactions with uranium or fission products.

‡Also from ternary fission.

NM: not measured

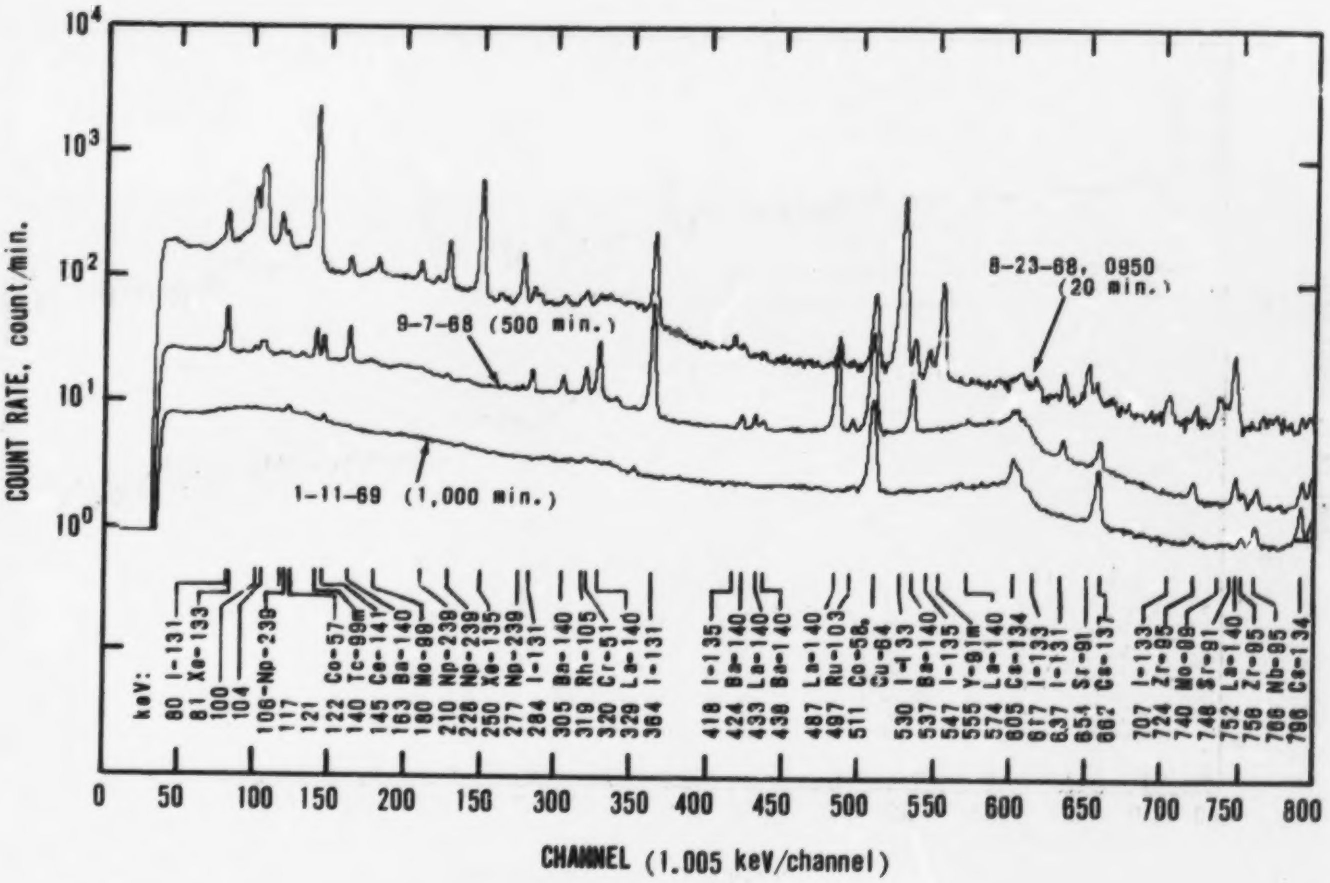


Figure 2.2. Gamma-ray spectra of primary coolant water, 40–804 keV. Detector: Ge(Li), 10.4 cm^2 × 11 mm, trapezoidal. Sample: 35 ml (including 1.7 ml acid) collected Aug. 22, 1968, 0710 CDT. (Counter background not subtracted)

water at pH 7.0. When this sample was filtered through Whatman #42 paper, between 10 and 50 percent of barium, curium, strontium, copper, cesium, and cobalt radionuclides, and less than 10 percent of all others from this sample were retained by the paper. The acidified sample lost smaller fractions of these radionuclides, but more radioiodine. Radiocolloidal behavior probably was responsible for radionuclide losses in the unacidified solution; the loss of one-third of the iodine from the acidified solution was attributed to the volatility of I_2. Radioiodine was therefore measured in unacidified samples, and radionuclides retained on container walls were collected by leaching with 3 portions of hot aqua regia.

2.3.3 *Generation rates of fission products in fuel and transfer rates to primary coolant.* Turnover rates of radionuclides in the primary coolant were computed from the measured concentrations to relate the latter to production rates in the fuel and release rates in effluents. The resulting ratios of rates of production to turnover, and turnover to release of radionuclides are quite approximate, as discussed below, but are simple and convenient indicators in a system that is extremely complex.

At the average power during operation of approximately 600 MWt in the period of interest, the generation rate, A, in μCi/sec, for each fission product was:

$$A = 600 \text{ MWt} \times \frac{3.1 \times 10^{16} \text{ fission}}{\text{MWt} \times \text{sec}} \times Y \times \lambda \text{ sec}^{-1} \times \frac{\mu\text{Ci} \times \text{sec}}{3.7 \times 10^4 \text{ dis}} \qquad (2.1)$$

where Y is the fission yield fraction and λ is the decay constant. Values of Y, λ,

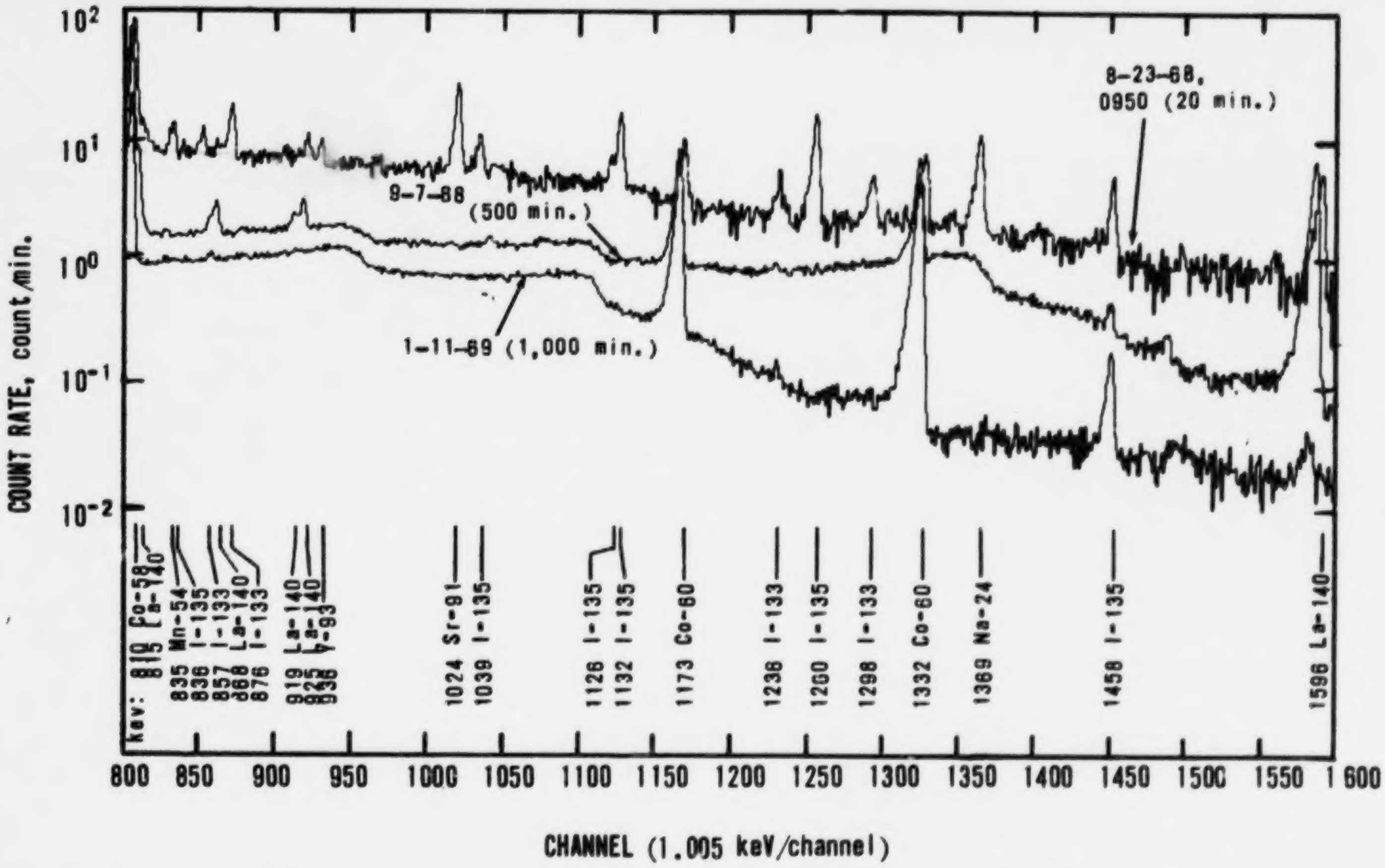


Figure 2.3. Gamma-ray spectra of primary coolant water, 805–1,608 keV. Same sample and detector as in Figure 2.2.

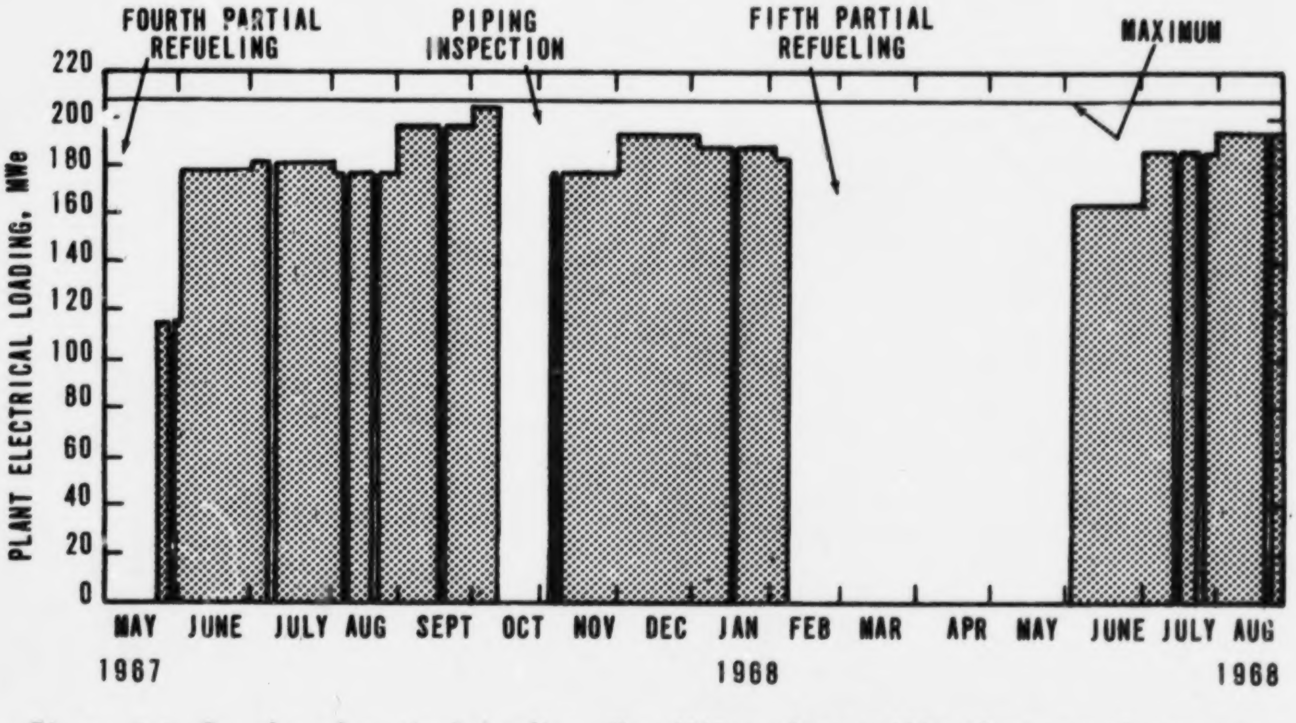


Figure 2.4. Dresden electrical loading, May 1967 to August 1968 (from annual reports of station operation for the years 1967 and 1968 by Dresden Nuclear Power Station).

and A are summarized in Appendix B.1. This is a simplification in that the calculation ignores (n,γ) reactions that link some fission-product chains, and production of some of these nuclides by activation of material in the coolant water (see Table 2.1). Moreover, the actual power varied considerably, and some values of Y are doubtful. Note that the concentration of a fission product in the fuel is proportional to A only if the half life is long compared to the irradiation time; for other fission products, relative concentrations are less because their radioactive decay rates approach or equal their generation rates.

The rate of turnover of fission products in the primary coolant, R, in μCi/sec, is related to the measured concentration, C, in μCi/ml, by:

$$R = C \frac{\mu Ci}{ml} \times V \text{ ml} \times (\lambda_{decay} + \lambda_{turnover}) \text{ sec}^{-1} \quad (2.2)$$

where V is the volume of coolant (1.9×10^8 ml) and $\lambda_{turnover}$ is the turnover constant for ions in the reactor-water demineralizer

$$\left(\frac{1}{14 \text{ hr} \times 3600 \text{ sec/hr}} = 2.0 \times 10^{-5} \text{ sec}^{-1}\right).$$

Values of R for the August 22 sample are given in Appendix B.2.

The turnover rate, R, can be taken to be equal to the rate of transfer from the fuel, except for the following limitations:

1. It is an instantaneous value and does not account for changes in concentration;
2. Other sources, such as release of radionuclides from ''crud'', are not considered;
3. The turnover rate, based on the flow rate through the reactor demineralizer shown in Figure 2.1, is appropriate to the time of sampling, but may vary from 10,000 to 50,000 kg/hr;(2) moreover, fission products that are not completely retained by the demineralizer have effective turnover times that exceed the turnover time of the water.
4. Turnover of insoluble and volatile fractions is not taken into account.

The ratios of turnover rates in the primary coolant to generation rates in fuel, R/A, (see Appendix B.2) range from the maximum 7.7×10^{-6} for ^{137}Cs to the minimum 1.5×10^{-9} for ^{132}Te. The radionuclides that are expected to be least soluble in the coolant water—Ce, Y, Nd, Zr, Nb, and Te—have the lowest ratios, below 4×10^{-8}. These radionuclides probably are mostly—i.e., 90 to 99.9 percent—in ''crud''. That longer-lived isotopes generally show higher R/A ratios is attributable to their greater accumulation in the fuel, which would result in higher transfer rates to the coolant.

2.3.4 *Generation rates of progeny of gaseous radionuclides in coolant.* The fission products ^{89}Sr, ^{90}Sr, ^{91}Sr, ^{137}Cs, and ^{140}Ba enter the primary coolant at least partially by decay of gaseous precursors in the coolant within the reactor. All are granddaughters of short-lived gaseous radionuclides with short-lived daughters except ^{137}Cs, which is the daughter of a short-lived gaseous radionuclide. The precursor gases enter the coolant from the fuel, decay partially in the reactor vessel, and then are carried in steam to the turbine.

Rates of formation of the descendants of these gaseous radionuclides within the reactor vessel could be computed if the transfer rate of the gases into the coolant and the fractional decay of the gases in the reactor vessel were known. The transfer rates of radioactive krypton and xenon have been observed to lie between two extremes. An ''equilibrium mixture'' (time of production and transfer at least one month so that all noble gas fission products except 10.7-yr ^{85}Kr have reached equilibrium) results from cladding with microscopic defects;(1) a ''recoil mixture'' (immediate transfer after formation) is associated with fuel that is in direct contact with coolant.(1) The intermediate state is termed ''diffusion mixture'' and has been taken(4,10) to be proportional to $\lambda^{0.5}$. According to current opinion, the intermediate state represents the combined effect of equilibrium and recoil situations, rather than a distinct transfer mechanism, although calculations based on bulk transfer mechanism predict the dependence on $\lambda^{0.5}$.(1)

At Dresden, the rate of transfer of each gaseous radionuclide from fuel, R_{dif}, in μCi/sec, has been related empirically to a nominal gaseous fission product release rate, N, in μCi/sec, by:(4)

$$R_{dif} = 4.0 \times 10^{2} \times Y \times \lambda^{0.5} \times N \qquad (2.3)$$

N is the sum of release rates computed for a diffusion mixture of the six radionuclides ^{85m}Kr, ^{87}Kr, ^{88}Kr, ^{133}Xe, ^{135}Xe, and ^{138}Xe, as shown in Appendix B.3. Dresden reports a value of N daily on the basis of a gamma-ray measurement of an off-gas sample.

The generation rate of the non-gaseous, relatively long-lived, progeny of the gases in the reactor vessel, R_p, in μCi/sec, is:

$$R_p = R_{dif(g)} \times f \times \lambda_p/\lambda_g \qquad (2.4)$$

where the subscripts p and g refer to progeny and gases, respectively, and f is the fraction of gaseous precursor decaying in the reactor vessel in approximately 10 seconds, as given for Dresden.(4) Values of R_{dif}, f, and λ_g are tabulated in Appendix B.3.

The generation rates (R_p), predicted coolant concentrations, and ratios of the generation rate from gas to the total transfer rate to the coolant based on concentration measurements for the August 22 sample (R values in Appendix B.2, N = 11,600 μCi/sec) are:

Radio-nuclide	R_p	Predicted concen-trations	R_p/R
^{89}Sr	0.027 μCi/sec	7×10^{-6} μCi/ml	0.16
^{90}Sr	0.00024	6×10^{-8}	0.049
^{91}Sr	6.0	8×10^{-4}	0.066
^{137}Cs	0.00018	5×10^{-8}	0.0011
^{140}Ba	0.14	4×10^{-5}	0.035

The small ratios of predicted to measured values suggest that, at Dresden, the recoil mechanism yields higher transfer rates to the coolant for these very short-lived gaseous precursors than is indicated by the diffusion mixture, or that decay of gaseous precursors in the coolant accounts for only a small fraction of these radionuclides in the coolant.

2.3.5 *Turnover rates of gaseous radioiodine.* Transfer rates of gaseous radioiodine into the primary coolant in a diffusion mixture also can be estimated with equation 2.3 and the values of Y and λ for ^{131}I, ^{133}I, and ^{135}I given in Appendix B.1. The estimated transfer rates and ratios of the estimated rates to turnover rates based on concentration measurements on August 22 (R value in Appendix B.2) are:

Radionuclide	R_{dif}	R_{dif}/R
^{131}I	130 μCi/sec	15
^{133}I	920	7.3
^{135}I	1,500	4.5.

That estimated rates were appreciably higher than measured rates suggests that the radioiodine does not pass from the fuel as rapidly as the noble gases. The possibility that most of the radioiodine does not remain in the aqueous phase was eliminated by the observation that only a small fraction of ^{131}I is in stack and liquid effluents (see Sections 3.3.3 and 4.3.3).

Ratios of turnover rates (computed according to equation 2.2) for the three radioiodine isotopes relative to ^{131}I compare as follows with theoretical ratios for the three types of mixtures:

	Measured Ratio		Predicted Ratio*		
Radionuclide	Feb. 1	Aug. 22	equilibrium	diffusion	recoil
^{131}I	1.0	1.0	1.0	1.0	1.0
^{133}I	12	14	2.2	6.8	21
^{135}I	39	38	2.1	11	60

*equilibrium rate = kY, where k is a constant
diffusion rate = $kY\lambda^{0.5}$
recoil rate = $kY\lambda$

The two sets of ratios of transfer rates computed from measured concentration agree with each other within the errors of measurement. They fall between ratios for diffusion and recoil mixtures, supporting the observation that the recoil mechanism may predominate.

2.3.6 *Formation and turnover rates of tritium.* The rate of turnover of tritium in the primary coolant, computed by equation 2.2 but using the turnover constant for demineralized water at Dresden instead of the turnover constant for ions in the reactor-water demineralizer, is

$$R = 1 \times 10^{-3} \frac{\mu Ci}{ml} \times 2 \times 10^{9}\ ml$$

$$\times (1.78 \times 10^{-9} + 2.8 \times 10^{-8})\ sec^{-1}$$

$$= 6 \times 10^{-2}\ \mu Ci/sec$$

The calculation is based on the presence of approximately 2×10^{6} liters of contaminated demineralized water and a release rate of 1.45×10^{5} liters per month at Dresden,(2) and an average tritium concentration of 1×10^{-3} μCi/ml, based on the values in Tables 2.1 and 2.2. The release rate is derived from the assumption that most demineralized water is released as high-conductivity liquid waste (i.e., release in stack effluent is relatively small), and that this waste is comprised mostly of contaminated demineralized water. The ratio of contaminated demineralized water on site to the release rate of high-conductivity liquid yields a turnover period of 420 days, hence, a turnover constant of 2.8×10^{-8} sec^{-1}.

Equation 2.3 was used to indicate R_{dif} for comparison with the turnover rate, although the rate of transfer from fuel thus calculated may be appreciably in error: tritium may leave the fuel more rapidly than other gases because of its smaller molecular size, but is too long-lived to have reached a constant transfer rate. The calculation yields, for ^{3}H from ternary fission:

$$R_{dif} = 4.0 \times 10^{2} \times 9.5 \times 10^{-5}$$

$$\times (1.78 \times 10^{-9})^{0.5} \times 11{,}600$$

$$= 0.02\ \mu Ci/sec$$

To place this value in perspective, the average observed release rate was one-fourth of the rate computed for R_{dif} of ^{85}Kr (see Table 4.2), whose half life is similar to that of tritium.

The D(n,γ)T reaction in coolant water within the reactor is probably the main source of tritium in the primary coolant.(8) The formation rate by this activation process is computed according to:

$$R_{act} = \phi \Sigma V \lambda \qquad (2.5)$$

based on the following values:

ϕ, average neutron flux(11)	3.1×10^{13} n/cm^{2} sec
Σ, macroscopic thermal neutron cross-section	
normal deuterium content of hydrogen	1.5×10^{-4} atom/atom
hydrogen content of water	0.11 g/g
water density at 290°C and 1,000 psi(12)	0.74 g/cm^{3}
water (non-void) fraction in core(13)	0.88
microscopic cross-section(14)	0.57×10^{-27} cm^{2}/atom

$$(\Sigma = 6.02 \times 10^{23} \times 1.5 \times 10^{-4} \times 0.11 \times 0.74 \times 0.88 \times 0.57 \times 10^{-27} = 3.7 \times 10^{-9}\ cm^{-1})$$

V, water volume in reactor core(11)	1.05×10^{7} cm^{3}
λ, tritium decay constant	1.78×10^{-9} sec^{-1}

Thus,

$$R_{act} = 3.1 \times 10^{13} \times 3.7 \times 10^{-9} \times 1.05 \times 10^{7} \times 1.78 \times 10^{-9} \times (3.7 \times 10^{4})^{-1}$$

$$= 0.06\ \mu Ci/sec$$

Most of the values used to compute the above rates are approximate; the deuterium content of the water would be expected to be higher than indicated because of the H(n,γ)D reaction, for example.

Although the sum of the two predicted rates, 0.02 + 0.06 = 0.08 μCi/sec, is not far from 0.06 μCi/sec based on water turnover, one or both of the predicted rates may be too high, since boron in the control rods may also be a source of tritium in primary coolant water.(8) The total rate, however, is confirmed by the measured release rate of tritium in liquid waste (0.05 μCi/sec, see Section 3.3.3).

2.3.7 *Turnover rate of activation products in primary coolant.* The turnover rates of activation product radionuclides with half lives longer than 1 week were computed with equation 2.2 and are listed in Appendix B.4. These rates were computed for comparison with release rates (see Sections 3.3.3 and 4.3.2). The rates apply only to the August 22, 1968 sample, and are subject to the same limitations discussed for fission products (see Section 2.3.3).

2.3.8 *Radionuclides in fuel pool and contaminated demineralized water.* Only long-lived radionuclides were found in these two samples (see Table 2.2). Tritium was the radionuclide at highest concentration in both; values were approximately two-thirds of the primary coolant concentration. Fuel pool water also contained relatively high concentrations of ^{134}Cs and ^{137}Cs, while contaminated demineralized water contained only traces of radionuclides other than tritium. The radionuclides ^{54}Mn, ^{90}Sr, ^{134}Cs, and ^{137}Cs were at higher concentration in the fuel pool than in the primary coolant sample collected at the same time. An analysis of contaminated demineralized water at Dresden in 1963 showed ^{64}Cu (17 x 10^{-6} μCi/ml) and also traces of ^{58}Co, ^{60}Co, ^{110m}Ag, and ^{137}Cs.(4)

Table 2.2

Radionuclide Concentrations in Fuel Pool Water and Contaminated Demineralized Water, August 22, 1968*

	Radionuclide Concentration, pCi/ml†	
Radionuclide	Fuel Pool Water	Demineralized Water
^{3}H	880	930
^{54}Mn	6	< 0.1
^{58}Co	18	1.2
^{60}Co	74	2.0
^{89}Sr	2	< 0.2
^{90}Sr	18	0.2
^{134}Cs	310	2.8
^{137}Cs	760	1.4
^{140}Ba	15	< 0.1
^{131}I	< 1	< 0.1
Gross β	~1,000	<10

*Concentration at time of sampling.

†1 pCi/ml = 1 x 10^{-6} μCi/ml.

Note: < values indicate minimum detectable activity at the ~3 σ confidence level.

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3. RADIONUCLIDES IN LIQUID WASTE EFFLUENTS

3.1 Samples

3.1.1 *Collection.* Samples of high-conductivity (high salt content) liquid wastes were collected by Dresden staff in 1-liter polyethylene containers from the 5000-gal waste tank(1) on November 12, 1967, January 13, 1968, March 16, 1968, June 25, 1968 and August 20, 1968. The liquid waste was circulated by pumping during sampling in accord with routine sample collection procedure at Dresden. Samples of laundry waste were collected by Dresden staff from the 1000-gal Laundry Waste Tank(1) on January 15 and March 12, 1968.

Radioactive ions were collected for analysis from the cooling-water intake and discharge canals by passing the water samples through ion-exchange columns as follows:

Date, 1968	Sample	Volume, liters	Collection period, hours	Hardness, as $CaCO_3$, mg/liter
Jan. 17–18	discharge	40	18	360
	intake	48	21.5	
June 25–26	discharge	65	16	160
	intake	65	16	
Aug. 20	discharge	153	1.5	200
	intake	131	1.5	

Intake water was pumped from the point of entrance into the plant; discharge water was pumped from the sampling station, approximately 100 m beyond the point of release of liquid wastes into the cooling-water discharge canal. For the first two sets of samples, water was passed continuously through the column during sampling; for the third set, water was first collected in 55-gal drums and then passed through the columns. The initial flow rate of 100 ml/min decreased gradually until the flow stopped in the first two sets of columns, but was maintained for the third set. The second and third samples were collected well within the 2-day periods during which high-conductivity waste was being discharged from its tank. The first sample was collected near the beginning of this period, and near the end of the period of release of another batch of liquid waste.(1)

One-liter aliquots of water were collected at the time of sampling to measure tritium, and hardness as $CaCO_3$. On November 15, 1967, additional samples were collected for tritium analysis in the Illinois River at Dresden Dam, 1.5 km downstream from the cooling-water canal discharge, and in the Kankakee River, 0.8 km upstream from the cooling-water canal intake.

3.1.2 *Liquid wastes.*(1) Almost all of the radioactivity released to the Illinois River by Dresden is in the high-conductivity liquid waste. The waste is a mixture from several sources including cleaning operations and laboratory discharges; the main constituent is spent solution (NaOH, H_2SO_4, and demineralized wash water) from regenerating the mixed-bed ion-exchange resin in the condensate demineralizer. The waste solution is at pH 11–12, and is filtered prior to collection in the liquid waste tank, so that insoluble radionuclides are transferred to solid radioactive waste. The waste sampled on August 20, however, was not filtered because the filter was broken.

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Liquid waste is discharged at rates up to 10 liters/min (2.7 gal/min), into the coolant-water discharge canal which has a flow rate of 630,000 liter/min (166,000 gal/min). The waste is usually discharged as the tank becomes full, but additional tanks are available if needed. The release rate is adjusted on the basis of gross beta activity to maintain gross radionuclide concentrations in the coolant canal below selected levels. At the usual release rates between 5 and 10 liters/min, this waste is discharged during less than one-half the operating time, and is diluted approximately 100,000-fold.

The average monthly volume of high-conductivity waste during 1968 was 145,000 liters (38,000 gal). Of the 5 analyzed samples, 4 were collected during reactor operation at power levels between 160 and 200 MWe (see Figure 2.4); the March 15 sample was collected during refueling.

The average monthly volume of laundry waste was 68,000 liters (18,000 gal) in 1968. It is released into the coolant-water discharge canal when it is necessary to empty the tank, at a flow rate of 10 liter/min.

3.1.3 *Effluent radioactivity*. Concentrations of radionuclides in effluents to unrestricted areas are limited by the AEC according to paragraph 20.106 of 10 CFR 20.(2) Concentrations above background in water, as listed in Appendix B, Table II, column 2 of 10 CFR 20, are applied at the boundary of the restricted area. The limit is 1×10^{-7} μCi/ml for an unidentified mixture containing no ^{129}I, ^{226}Ra, or ^{228}Ra; and 3×10^{-7} μCi/ml for soluble ^{90}Sr or ^{131}I, which are usually the radionuclides in reactor effluent with the lowest limits. Higher limits are permissible under conditions of subsection (b), or more stringent limits may be applied by the AEC under subsection (e) of paragraph 20.106.

At Dresden, concentrations of released radionuclides are determined by measuring gross beta concentrations in liquid waste tanks and applying the dilution factor (the ratio of canal flow rate to tank discharge rate) appropriate to the point of release. In 1968, "the average contribution to the unidentified activity in the water utilized for radioactive waste dilution during the year was calculated to be 0.189×10^{-7} μCi/ml" at Dresden.(3) Concentrations during the preceding five years were within a factor of two of this value, according to Dresden's annual reports for these years.

3.1.4 *Effluent sampler*. An apparatus was designed, tested and used at Dresden on the three occasions listed in Section 3.1.1 to collect radionuclides in ionic form continuously from water at the point of release. A minimum sample volume of 10 liters was considered necessary to obtain a sensitivity of 1×10^{-9} μCi/ml (1 pCi/liter) for each radionuclide that was known to be discharged. The tested device processed as much as 150 liters of water.

The apparatus consists of five sections of ion-exchange resin in a 7.5-cm i.d. column. Each of the first three sections contains 350 cc Dowex* 50-x8 (H^+, 50-100 mesh) cation-exchange resin; each of the fourth and fifth sections, 450 cc Dowex 1-x8 (Cl^-, 50-100 mesh) anion-exchange resin. The sections are separated by porous plastic sheets so that they can be taken separately from the column. A glass wool filter precedes the first section. The apparatus was tested in the laboratory with tracer-level radio-cesium, -strontium, and -cobalt in 150 liters of water with a hardness of 200 mg/liter at a flow rate of 100 ml/min. More than 90 percent of each radionuclide was retained by the first cation-exchange resin section, and more than 99 percent by the first and second in combination. More than 90 percent of radioiodine tracer was retained by the upper anion-exchange resin section from water that did not contain oxidizing agents such as chlorine. To analyze chlorinated water (i.e., drinking water), sufficient $NaHSO_3$ was added to keep ^{131}I reduced as iodide. Thus, under appropriate conditions, these tested radionuclides can be collected almost completely in the upper sections, and the appearance of large fractions in lower sections is a warning of breakthrough.

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*Mention of commercial products does not constitute endorsement by the Public Health Service.

3.2 Analysis

3.2.1 *Liquid wastes.* Samples were usually divided into two portions—one unfiltered and one filtered (the second and third samples through Whatman #42 paper, the fourth and fifth, through a membrane filter)—for analysis. The high-conductivity waste of November 12 and both laundry wastes, however, were not filtered. Ten-ml aliquots were evaporated to dryness on planchets to determine gross-beta concentrations with low-background (1 count/min) beta-particle counters, and gross-alpha content with proportional counters. Radionuclide analyses were performed by gamma-ray spectrometry with Ge(Li) and NaI(Tl) detectors, and by chemical separation and subsequent counting as described in Section 2.2. All radionuclides observed by gamma-ray spectrometry (see Figure 3.1) had half lives longer than 1 week, hence chemical analysis was restricted to the longer-lived radionuclides that are expected to be in these samples. Concentrations were measured within ± 1 pCi/ml for high-conductivity waste, and ± 0.1 pCi/ml for laundry waste.

3.2.2 *Coolant-canal samples.* The five sections of ion-exchange resin in each column were separated for analysis, and each section was washed with distilled water to remove silt. Each section was analyzed spectrometrically with a 10 X 10 cm cylindrical NaI(Tl) gamma-ray detector, as shown in Figures 3.2 (top cation-exchange resin) and 3.3 (upper anion-exchange resin). Anion-exchange resin sections that appeared to contain ^{131}I were measured again after 1-week intervals so that gamma spectral identification could be confirmed by the decay rate of the gamma-ray peak. Each cation-exchange resin section was placed separately into a small column and the cations were eluted from the resin with 1.2 liter 6 N HCl. Aliquots of the elutriant were analyzed for radio-cesium, -strontium,

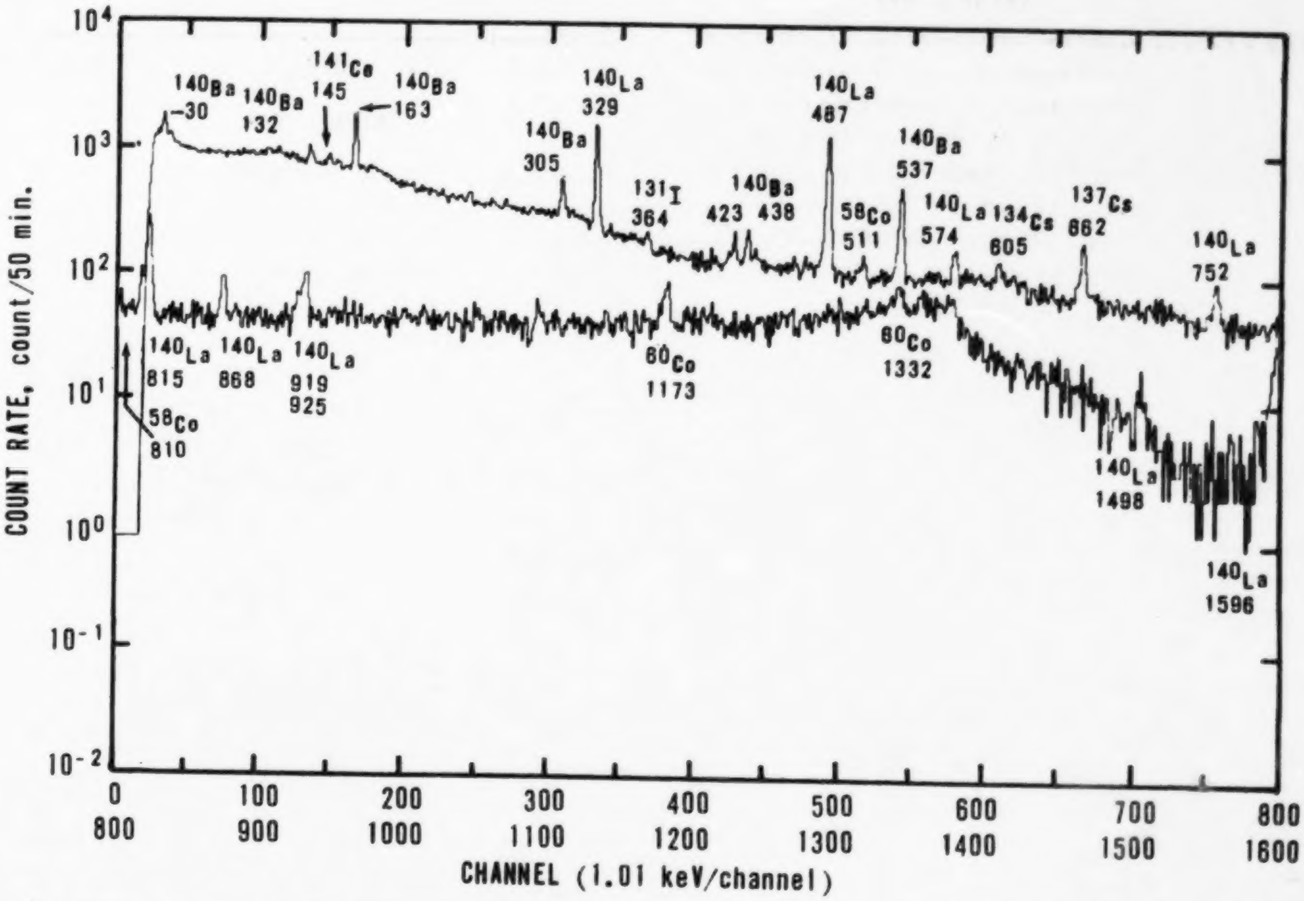

Figure 3.1. Gamma-ray spectrum of high-conductivity waste solution. Detector: Ge(Li), 6 cm^2 x 7 mm. Sample: 400 ml collected Jan. 13, 1968. Count: Jan. 19 (50 min.).

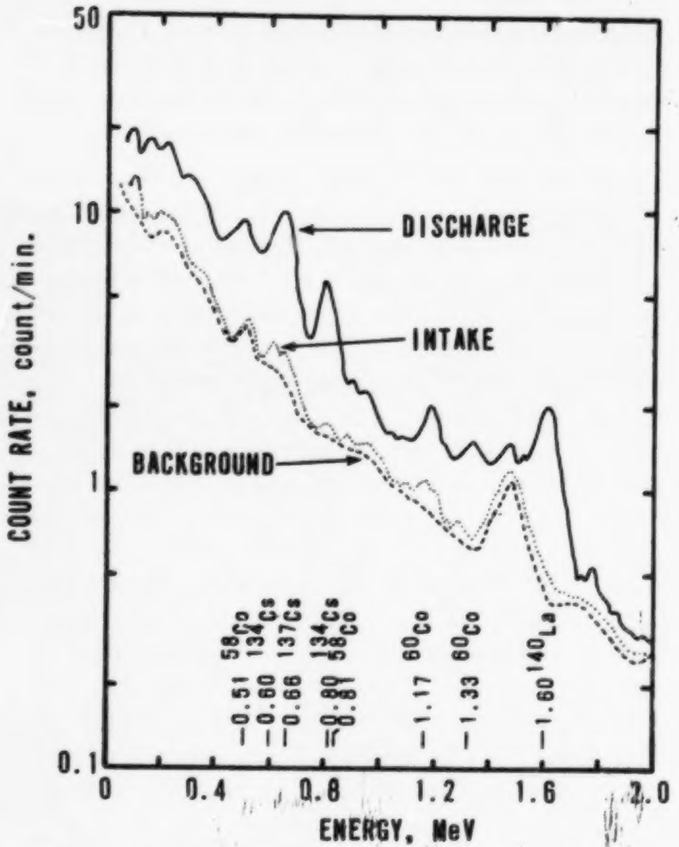

Figure 3.2. Gamma-ray spectra of cation-exchange resins in intake and discharge coolant-water canals. Detector: 10 × 10 cm NaI(Tl). Samples: 350 cc resin at top of columns used on Jan. 17–18, 1968. Counts: Jan. 21 (100 min.).

-cobalt, and -barium by chemical separation, and the separated samples were counted with a beta-particle detector or a gamma-ray spectrometer, as appropriate.

Silt washed from the resin sections was combined with silt retained by the glass wool filter, dried at 100 °C, and weighed. It was analyzed by gamma-ray spectrometry with the NaI(Tl) detector, and also by coincidence/anticoincidence spectrometry with two 10 x 10-cm NaI(Tl) detectors within a NaI(Tl) annulus. Examples of these spectra are given in Figures 3.4 and 3.5. Gamma-ray coincidence measurements enhance sensitivity for detecting ^{58}Co, ^{60}Co, and ^{134}Cs. Radiostrontium was determined in the silt samples by leaching 1-g aliquots with hot 1 N HNO$_3$, chemically separating strontium, and counting the total strontium

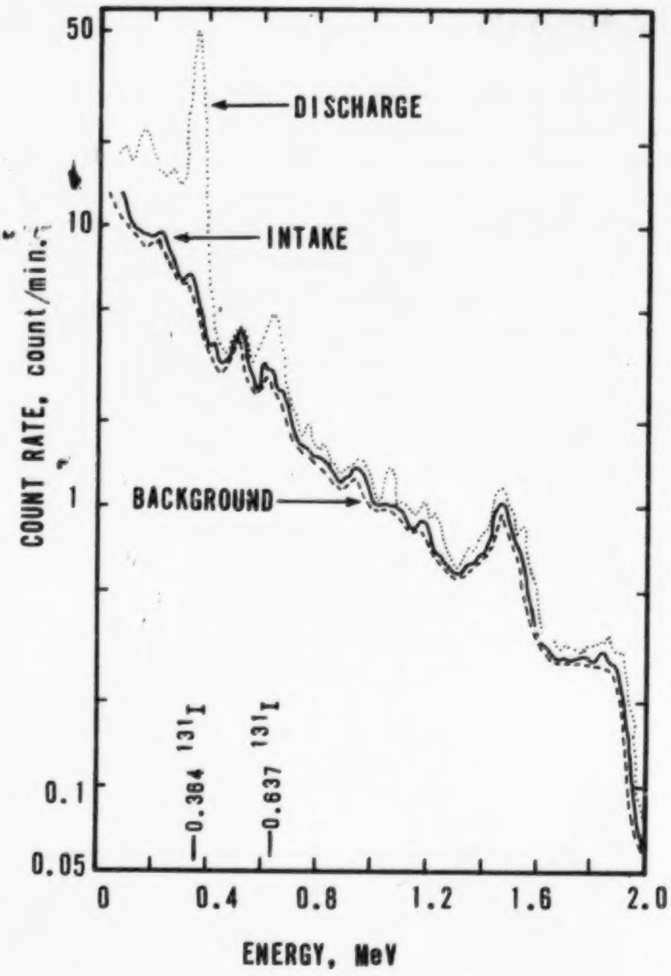

Figure 3.3. Gamma-ray spectra of anion-exchange resins in intake and discharge coolant-water canals. Detector: 10 × 10 cm NaI(Tl). Sample: 450 cc resin at top of anion-exchange section of columns used on Jan. 17–18, 1968. Counts: Jan. 21 (100 min.).

(^{89}Sr + ^{90}Sr) activity. The ^{90}Y daughter of ^{90}Sr was chemically separated after ingrowth and counted. The amounts of ^{89}Sr and ^{90}Sr were then computed from the count rates of total strontium and ^{90}Y.

The volume of water that had passed through the columns was determined by measuring the hardness in the water sample in terms of calcium carbonate (0.40 mg Ca^{++} = 1.00 mg hardness), and the hardness in the acid elutriant from all three sections of cation-exchange resin. In terms of calcium ion,

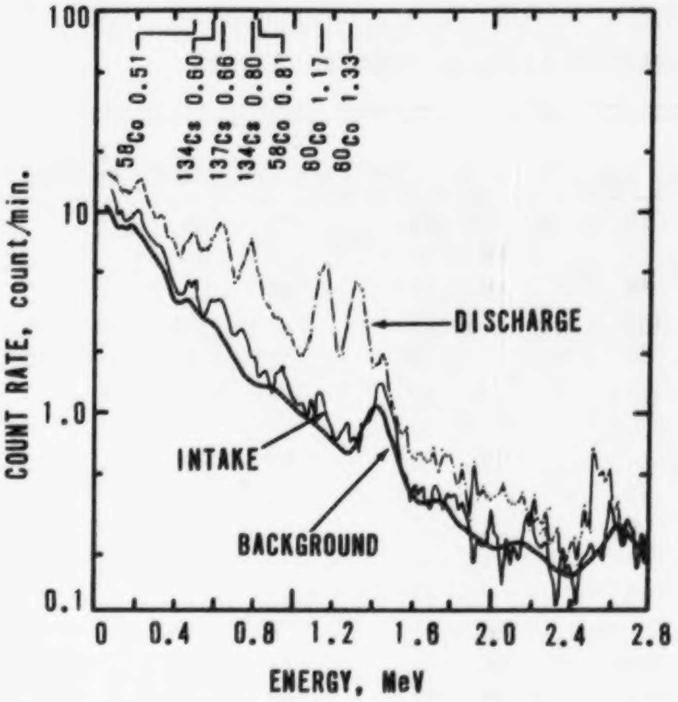

Figure 3.4. Gamma-ray spectra of suspended silt in intake and discharge coolant-water canals. Detector: 10 × 10 cm NaI(Tl). Samples: 3.2 g dried silt from intake and 4.4 g dried silt from discharge collected Aug. 20, 1968 at 1630–1800 CDT. Counts: Sept. 9 (100 min.).

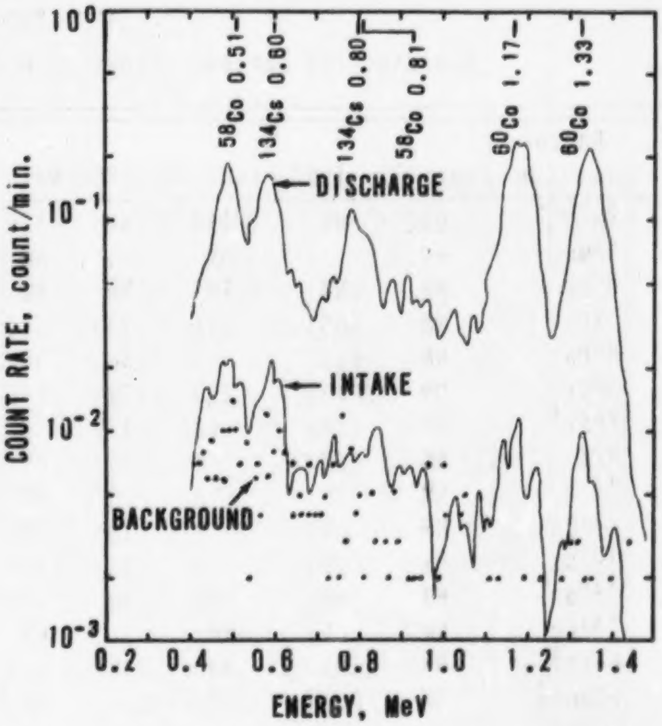

Figure 3.5. Gamma-ray coincidence spectra of suspended silt in intake and discharge canals. Detectors: Two 10 × 10 cm NaI(Tl) in coincidence, within 5-cm thick annulus in anti-coincidence. Samples: Same as in Figure 3.4. Counts: Nov. 19, 1968 (1,000 min.).

$$\text{Liters of water} = \frac{\text{total calcium eluted from resin}}{\text{calcium/liter of water sample}} \qquad (3.1)$$

In the third set of samples, the computed volume was confirmed by direct measurement.

The tritium content of the water samples was determined by distilling the water and analyzing 5-ml samples of the distilled water with a liquid scintillation counter that had been adapted to measure concentrations as low as 0.2 pCi/ml*.

3.3 Results and Discussion

3.3.1 *Radionuclide content of liquid wastes.* The average concentration of all detected radionuclides in the five samples of high-conductivity waste was approximately 2×10^{-3} μCi/ml, as shown in Table 3.1. The radioactive constituents at highest concentrations were ^{3}H, ^{58}Co, ^{60}Co, ^{89}Sr, ^{131}I, ^{134}Cs, ^{137}Cs, and ^{140}Ba. In addition to the listed radionuclides, traces of ^{35}S, ^{57}Co, ^{65}Zn, ^{95}Zr ^{95}Nb, ^{106}Ru, and ^{141}Ce were observed in one or more samples. Similar concentrations of the major radioactive constituents were observed in samples collected in 1963.(4)

The total activity and the concentrations of individual radionuclides varied considerably among samples. Average values were used to compute release rates of individual radionuclides only because better data (i.e., values measured throughout the year) are not available. To evaluate the causes of variability, it would be necessary to segregate waste liquids according

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*We thank Dr. A. A. Moghissi, Southeastern Radiological Health Laboratory (SERHL), BRH, EHS, PHS, DHEW, for analyzing these samples.

Table 3.1

Radionuclide Concentrations in High-conductivity Liquid Waste, pCi/ml*

Radio-nuclide	Nov. 12, 1967	Jan. 13, 1968	March 16, 1968	June 25, 1968	Aug. 20, 1968	Average
^{3}H	900** (NM)	950 (NM)	520 (NM)	770 (NM)	1,100 (NM)	850
^{54}Mn	NM (< 1)**	NM (< 1)	NM (< 1)	NM (< 1)	< 1 (22)	(4)
^{55}Fe	NM (NM)	NM (NM)	NM (NM)	NM (45)	NM (50)	(48)
^{58}Co	NM (65)	4 (34)	880 (1,800)	3 (85)	18 (260)	230 (450)
^{60}Co	NM (33)	1 (46)	500 (1,350)	4 (180	11(1,400)	130 (600)
^{89}Sr	NM (140)	220 (320)	140 (170)	85 (89)	24 (34)	120 (150)
^{90}Sr†	NM (14)	12 (17)	30 (30)	8 (9)	9 (11)	15 (16)
^{91}Y	NM (66)	1 (26)	NM (< 1)	NM (1)	NM (< 1)	(19)
^{131}I	NM (5)	6 (6)	NM (45)	46 (49)	15 (15)	22 (24)
^{134}Cs	NM (9)	7 (10)	80 (90)	47 (50)	27 (34)	40 (39)
^{137}Cs	NM (29)	35 (35)	150 (170)	140(160)	99 (103)	106 (99)
^{140}Ba†	NM (45)	40 (160)	65 (95)	22 (54)	35 (105)	41 (92)
^{144}Ce	NM (5)	NM (< 1)	NM (< 1)	NM (< 1)	8 (16)	(4)
beta‡	NM (540)	440(1,040)	1,600 (NM)	NM (550)	140 (NM)	
alpha‡	NM (< 0.1)	NM (< 0.1)	NM (NM)	NM (NM)	NM (NM)	

*concentration of radionuclides at indicated date of sampling; 1 pCi/ml = 1 x 10^{-6} μCi/ml.

**values without parentheses are for filtered solution, values in parentheses for unfiltered solution.

†solutions also contained ^{90}Y at concentrations equal to its ^{90}Sr parent; and ^{140}La at concentrations 1.15 times those of its ^{140}Ba parent.

‡gross radioactivity measured approximately 1 week after sample collection and not corrected for decay.

NM: not measured; < values are 3 σ counting error.

to origin. It may be noted that radionuclide concentrations were relatively high during refueling (March 16 sample), and that the tritium concentration in this sample was only two-fold lower than during reactor operation. The sample of June 25 may also have contained waste from refueling.

Gross beta and tritium values in laundry wastes (Table 3.2) were two orders of magnitude lower than in the high-conductivity wastes. Concentrations of ^{3}H, ^{58}Co, ^{60}Co, ^{89}Sr, ^{134}Cs, and ^{137}Cs were all approximately 1 x 10^{-6} μCi/ml.

3.3.2 *Solubility of radionuclides in high-conductivity waste.* As indicated in Table 3.1, the radionuclides in the sample were partially removed by filtering. The unfiltered waste of August 20 had the highest fraction of insoluble radionuclides, but even wastes that had been filtered prior to sampling contained some insoluble radionuclides. Radio-cesium, -strontium, -iodine, and -barium are in the filtered waste because they are relatively soluble in alkaline solutions. Radio-yttrium and -lanthanum probably were formed after filtration by decay of their radioactive precursors. Most other relatively insoluble radionuclides may represent traces of solids not retained by the filter. Radiocobalt appears to be dissolved in complex form initially, but becomes progressively less soluble on standing. In tests with ion-exchange resins, 90 percent of the soluble radiocobalt was retained on cation-exchange resins and 5 percent was retained on anion-exchange resins.

3.3.3 *Turnover rate in primary coolant vs. release rate in liquid waste.* Average release rates of radionuclides in the liquid waste, E, were computed from the average concentrations in Table 3.1 for comparison

Table 3.2

Radionuclide Concentrations in Liquid Laundry Waste,* pCi/ml

Radionuclide	January 15, 1968	March 12, 1968
^{3}H	1.3	1.5
^{58}Co	2.2	1.2
^{60}Co	2.3	0.6
^{89}Sr	< 0.1**	0.9
^{90}Sr	0.1	< 0.1
^{131}I	< 0.1	< 0.1
^{134}Cs	0.6	< 0.1
^{137}Cs	3.4	0.8
^{140}Ba	< 0.1	< 0.1
^{141}Ce	< 0.1	< 0.1
gross beta	10	5
gross alpha	< 0.1	< 0.1

*Radionuclide concentrations at sampling date; gross values approximately 1 week later.

**< values are 3 σ counting error.

with their turnover rates, R, in the primary coolant. The average concentrations (in pCi/ml) were multiplied by 56 x 10^{-6} to obtain release rates (in μCi/sec) at an average monthly discharge of 145,000 liters of high-conductivity effluent. These release rates are tabulated below with the ratio E/R (see Appendices B.2 and B.4 for values of R):

Radionuclide	Release rate, E	E/R
^{3}H	4.8 x 10^{-2} μCi/sec	---
^{54}Mn	2. x 10^{-4}	0.03
^{55}Fe	2.7 x 10^{-3}	0.018
^{58}Co	2.5 x 10^{-2}	0.0038
^{60}Co	3.4 x 10^{-2}	0.034
^{89}Sr	8.4 x 10^{-3}	0.049
^{90}Sr	9.0 x 10^{-4}	0.18
^{131}I	1.3 x 10^{-3}	0.0016
^{134}Cs	2.2 x 10^{-3}	0.025
^{137}Cs	5.5 x 10^{-3}	0.032
^{140}Ba	5.2 x 10^{-3}	0.0013
^{144}Ce	2. x 10^{-4}	0.007

These E/R ratios suggest that only small fractions of most longer-lived radionuclides measured in primary coolant water at the steam drum are released in liquid waste. Moreover, this water is not the only source of radionuclides. Some ^{89}Sr, ^{90}Sr, ^{137}Cs, and ^{140}Ba in the waste liquid undoubtedly were formed by decay of gaseous precursors in the turbine, condenser, and delay line; some of the longer-lived radionuclides, especially strontium and cesium, may be from the fuel pool (see Table 2.2,. For shorter-lived radionuclides, the ratio E/R was too small to measure. The release rate of tritium, on the other hand, is of the same magnitude as the turnover rates of 6 x 10^{-2} or 8 x 10^{-2} μCi/sec computed in Section 2.3.6 for contaminated demineralized water, hence the ratio E/R appears to be near unity.

3.3.4 *Concentration of ionic radionuclides at point of discharge.* The ionic radionuclides at highest concentration in the high-conductivity waste (see Table 3.1) were determined in coolant-canal water on three occasions after concentrating them on the ion-exchange device. Coolant water from the intake canal contained only the radionuclides ^{90}Sr and ^{137}Cs at average concentrations of 0.4 and 0.2 x 10^{-9} μCi/ml, respectively, according to Table 3.3; these were undoubtedly deposited by fallout from atmospheric nuclear weapon tests. The difference in radionuclide concentrations between water in the coolant-water discharge and intake canals is attributed to radionuclide releases at Dresden. These values are considerably different from estimates based on liquid waste releases at the time of sample collection. The sums of measured radionuclide concentrations near the point of discharge (in Table 3.3) compare as follows to estimates based on gross beta values (or summed radionuclide concentrations, excluding ^{3}H, for the August 20 sample) in high-conductivity liquid waste:

Date	Dilution factor in canal	Measured total beta from Table 3.3	Estimated gross beta from Table 3.1
January 17–18	62,000	39 pCi/liter	17 pCi/liter
June 25–26	90,000	2	6
August 20	100,000	1	21

Similarly large differences exist for individual radionuclides.

The higher concentrations measured in the coolant-water discharge canal on January

Table 3.3

Concentration of Ionic Radionuclides in Coolant-canal Water, pCi/liter*

Radio-nuclide	Sample	Jan. 17–18, 1968†	June 25–26, 1968	Aug. 20, 1968
^{58}Co	discharge	2.7	< 0.3	0.2
	intake	< 0.1	< 0.3	< 0.1
^{60}Co	discharge	1.0	< 0.3	0.2
	intake	< 0.1	< 0.3	< 0.1
^{89}Sr	discharge	7.5 ± 0.5	< 0.1	< 0.2
	intake	< 0.1	< 0.1	< 0.2
^{90}Sr	discharge	0.8	0.5	0.5
	intake	0.5	0.4	0.3
^{131}I	discharge	17. ± 2	0.8 ± 0.3	< 0.2
	intake	< 0.2	< 0.2	< 0.2
^{134}Cs	discharge	1.6	< 0.2	< 0.1
	intake	< 0.1	< 0.2	< 0.1
^{137}Cs	discharge	4.3	0.2	0.2
	intake	0.3	0.1	0.1
^{140}Ba	discharge	3.8	0.6 ± 0.3	0.2
	intake	0.1	< 0.3	< 0.1

*concentration at date of sampling; 1 pCi/liter = 1 x 10^{-9} μCi/ml.

†silt not separated from ion-exchange resin in this sample.

Note: 2 σ counting errors are ± 0.1 pCi/liter or as shown; < values are 3 σ counting errors.

17–18 were traced to an unusual liquid waste—blowdown water from the secondary steam generators to which high-conductivity water had inadvertently been added—that was released just before the one described in Table 3.1.(1) The lower measured concentrations in the two other samples can be ascribed to radionuclides that were insoluble or retained by silt, since these are not collected on the ion-exchange resins. Approximately one-half of the radioactivity other than tritium was insoluble in the June 24–25 sample, and 90 percent in the August 20 sample (see Table 3.1). Concentrations of most soluble radionuclides in the liquid waste sampled on August 20, divided by the applicable dilution factor, compare reasonably well with the measured differences in concentration between coolant canal discharge and intake water:

Radionuclide	Concentration of soluble fraction in liquid waste ÷ 100,000	Concentration at point of discharge*
^{58}Co	0.18 pCi/liter	0.2 ± 0.1 pCi/liter
^{60}Co	0.11	0.2 ± 0.1
^{89}Sr	0.24	< 0.2
^{90}Sr	0.09	0.2 ± 0.1
^{131}I	0.15	< 0.2
^{134}Cs	0.27	< 0.1
^{137}Cs	0.99	0.1 ± 0.1
^{140}Ba	0.35	0.2 ± 0.1

*Concentration of radionuclides in coolant-canal water at point of intake has been subtracted.

Measured radiocesium values, however, were appreciably lower at the point of discharge than estimated from analysis of the liquid waste in the tank.

For comparison, gross beta measurements of composite canal-water samples by the contractor for environmental surveillance at Dresden during the first and second sampling periods are as follows:(5)

Date	Location	Gross beta concentration
Jan. 15–21, 1968	Discharge canal Intake canal	6.8 ± 0.8 pCi/liter 3.9 ± 0.4
June 23–29, 1968	Discharge canal Intake canal	16.4 ± 1.6 14.0 ± 1.2

These values were probably obtained for samples that had been evaporated and thus did not retain ^{3}H and other volatile radionuclides. The weekly sampling period may include several discharges as well as intervals during which no discharge occurred. The highest difference in weekly gross beta concentrations between discharge and intake canals in the first 6 months of 1968 was 13 pCi/liter.(5)

3.3.5 *Retention efficiency of effluent sampler*. The sample of January 17–18 had the highest radionuclide concentration of the three, and therefore provided the best comparison of retention on the successive ion-exchange resin sections. Values for the column in the discharge canal, through which 40 liters of water had been passed, were as follows in terms of pCi/liter influent water:

Substance	Section 1	Section 2	Section 3	Total
^{58}Co	2.7	< 0.4	< 0.3	2.7
^{60}Co	1.0	< 0.3	< 0.2	1.0
^{89}Sr	7.5	< 0.1	< 0.1	7.5
^{90}Sr	0.8	< 0.1	< 0.1	0.8
^{131}I	17.	< 0.2	---	17.
^{134}Cs	1.6	< 0.2	< 0.2	1.6
^{137}Cs	4.0	0.3	< 0.1	4.3
^{140}Ba	3.8	< 0.4	< 0.4	3.8
Ca^{++} (mg/liter)	120	0.7	0.4	121

The section number refers to anion-exchange resin for ^{131}I, and cation-exchange resin for all others. The values indicate that 90 percent or more of each radionuclide and 99 percent of calcium was retained in the top column. Such confirmation is important because either interfering substances or a difference in the chemical form of the ion could reduce the retention efficiency demonstrated in tracer studies.

3.3.6 *Insoluble radionuclides suspended in cooling-canal water*. The amounts of radionuclides washed with distilled water from the glass wool filter and ion-exchange resins are given in Table 3.4. Concentrations in water were computed by multiplying the radionuclide content in the silt by the grams of silt collected per liter of water passing through the columns. As in the case of ionic radionuclides, the ^{90}Sr and ^{137}Cs in the intake canal are assumed to be from fallout, and differences between discharge- and intake-canal values, from Dresden. Traces of ^{89}Sr and ^{60}Co in the intake water may indicate minor contamination in the intake canal at a time prior to sampling, or the ^{89}Sr may have been from fallout. In addition, the gamma spectra (Figures 3.4 and 3.5) show the naturally occurring radionuclides ^{40}K, ^{226}Ra plus progeny, and ^{232}Th plus progeny in silt from the intake and discharge canals.

The suspended radioactivity was mostly radiocobalt and radiocesium, and included some radiostrontium and ^{140}Ba. A considerable fraction of the radiocobalt and radiocesium in the coolant-water discharge canal was associated with suspended material, as indicated by comparing Table 3.4 with Table 3.3. Radiocobalt probably was collected with suspended material because much of it was insoluble in the discharged waste (see Table 3.1). Radiocesium, although entirely soluble in the waste tank, was probably retained partially in silt suspended in coolant-canal water. Two-thirds of the soluble radiocesium and 95 percent of the insoluble radiocobalt from the liquid waste of August 20 remain unaccounted for. These fractions may have been deposited in the canal upstream from the point of sampling, or suspended in material that was not collected by the sampler. An analysis of mud samples from the discharge canal in

Table 3.4

Radionuclide Content of Suspended Silt Collected from Coolant-canal Water

Radio-nuclide	Sample	June 25-26, 1968*		August 20, 1968*	
		pCi/g dried silt	pCi/liter water	pCi/g dried silt	pCi/liter water
^{58}Co	discharge	0.9	0.4	5 ± 1	0.1
	intake	< 0.1	---	< 1	---
^{60}Co	discharge	0.6	0.2	27 ± 1	0.7
	intake	< 0.1	---	1	0.02
^{89}Sr	discharge	0.1	0.04	< 3	---
	intake	0.1	0.02	< 3	---
^{90}Sr	discharge	0.1	0.04	1.3	0.04
	intake	0.1	0.02	0.2	0.005
^{131}I	discharge	< 0.1	---	< 0.3	---
	intake	< 0.3	---	< 0.3	---
^{134}Cs	discharge	< 0.2	---	3.5 ± 0.3	0.1
	intake	< 0.2	---	< 0.3	---
^{137}Cs	discharge	1.1	0.5	11 ± 1	0.3
	intake	0.8	0.1	3	0.07
^{140}Ba	discharge	< 0.5	---	5 ± 2	0.1
	intake	< 1.0	---	< 5	---

*Amounts of dried silt: June 25-26 discharge - 27.6 g/65 liter; intake - 11.1 g/65 liter.
Aug. 20 discharge - 4.4 g/153 liter; intake - 3.2 g/131 liter.

Note: 2 σ counting errors are ± 0.2 pCi/g or as shown; < values are 3 σ counting errors.

1963 showed 0.3 pCi ^{60}Co and 0.6 pCi ^{137}Cs per gram, as well as some radionuclides attributable to fallout.(4)

3.3.7 *Tritium concentrations in water.* Concentrations of tritium measured on three occasions were not significantly higher in the cooling-water discharge canal than in the intake or in other nearby samples of river water, as shown in Table 3.5. It is improbable that the difference of 0.4 pCi/ml in the two earlier samples is significant and attributable to Dresden. The concentration at the point of release estimated from the average tritium concentration of 850 pCi/ml on liquid waste (Table 3.1) at a dilution factor of 1 x 10^5 is 0.008 pCi/ml during releases of high-conductivity liquid wastes.

Table 3.5

Tritium Concentrations in Water, pCi/ml*

Location	Nov. 15, 1967	Jan. 17, 1968	June 25, 1968	Aug. 20, 1968
Cooling-water discharge canal	1.2 ± 0.3**	0.6 ± 0.3	< 1.5	0.2 ± 0.2
Cooling-water intake canal	0.8 ± 0.3	0.2 ± 0.2	< 1.5	0.2 ± 0.2
Illinois R. at Dresden Dam	0.8 ± 0.3	---	---	---
Kankakee R., 0.8 km upstream from cooling-water intake	1.0 ± 0.3	---	---	---

*Analysis by SERHL except for June 25 sample.

**± values are 2 σ counting error; < values are 3 σ counting error.

3.4 References

1. Kiedaisch, W. and R. Pavlick, Dresden Nuclear Power Station, 1968, personal communication.
2. U.S. Atomic Energy Commission, ''Standards for Protection Against Radiation'', Title 10, Code of Federal Regulations, Part 20 (U.S. Gov't. Printing Office, Washington, D. C.. 1965).
3. Dresden Nuclear Power Station, ''Annual Report of Station Operation for the Year 1968'' (Commonwealth Edison Co., Chicago, 1969), p. 21.
4. Gilbert, R. S., ''Sources and Disposition of Radioactive Liquid and Gaseous Effluents from the Dresden Plant'', General Electric Co. Rept. APED-4461 (1964).
5. Fried, R. E. and J. M. Matuszek, Jr., ''Environs Monitoring Program, Dresden Nuclear Power Station'', First and Second Quarterly Reports, 1968 (Isotopes, A Teledyne Company, 1968).

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4. RADIONUCLIDES RELEASED FROM STACK

4.1 Samples

4.1.1 *Description of system.* Boiling water reactors such as Dresden I continuously produce radioactive gases and aerosols and release them to the atmosphere during reactor operation. The discharged radionuclides at highest concentration are the fission-produced noble gases and ^{13}N. Their transfer within the plant and from source to stack is depicted schematically in Figure 4.1, which is based on descriptions from several sources.(1-4)

The main flow of gaseous radionuclides is from degassing of condensed steam by the air ejectors. The off-gas passes at a nominal flow rate of 9.4 l/sec through the delay line that provides a nominal 20-minute* period for removal by decay of most of the initial activity (see Appendix B.3). The decay period is especially important in reducing the release rates of 3.2-min ^{89}Kr, and 33-sec ^{90}Kr, the precursors of ^{89}Sr and ^{90}Sr, respectively. Before the gas is released to the base of the exhaust stack, it passes through high efficiency particulate air (HEPA) filters (nominal removal efficiency 99.97 percent at 0.3 μ size) to remove accompanying aerosols, especially the recently formed radioactive progeny of the noble gas fission products. An approximately 2250:1 dilution of the off-gas is provided at the stack base by ventilation air that is exhausted without filtration from the reactor containment structure and the turbine building.

Several secondary sources contribute radionuclides to the stack effluent. Approximately 0.1 percent of the process gas escapes through the turbine gland seals. This gas is separated from steam by a condenser and exhausted without filtration through a 2-minute delay line to the stack.(1) Ventilating air carries traces of radioactivity. Airborne radioactivity may also arise from minor leaks in the reactor pipeways and secondary steam generators.(3)

The 91-m-high exhaust stack diminishes from a base diameter of 3 m to 1.4 m at the top. The exhaust air requires 20 seconds for transit through the stack at the nominal exit flow rate of 21 m^3/sec. At the 30-m elevation (~ 10 sec. above the base), an 8-nozzle probe withdraws samples of the effluent for the stack monitoring system.(4) This air is pumped at a rate of 1.1 to 1.4 m^3/hr and passes sequentially through a 5-cm-dia. membrane filter for particulate collection, a 5-cm-dia. organic vapor respirator cartridge (27 g charcoal bed, Cesco type B) for collecting gaseous iodine, and a radiation detector for continuous on-line monitoring of noble gas activity. The filter and cartridge are routinely replaced at 24-hr intervals.

The stack discharge limit is prescribed by the AEC license at an annual average release rate for noble fission gases of 700,000 μCi/sec. This release rate is set in accord with 10 CFR 20 paragraph 20.105, to prevent exceeding a dose to the whole

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*DNPS staff reported recently that the actual flow rate is 10.4 l/sec (22 cfm), resulting in a holdup time of approximately 23 minutes and dilution factor of 2050:1. On this basis, predicted concentrations of noble gases and ^{13}N in the stack would be 9% higher due to less dilution. On the other hand, the holdup time would be 3 minutes longer, decreasing levels of the short half-life nuclides, 17-min. ^{138}Xe and 10-min. ^{13}N, by 11% and 19%, respectively.

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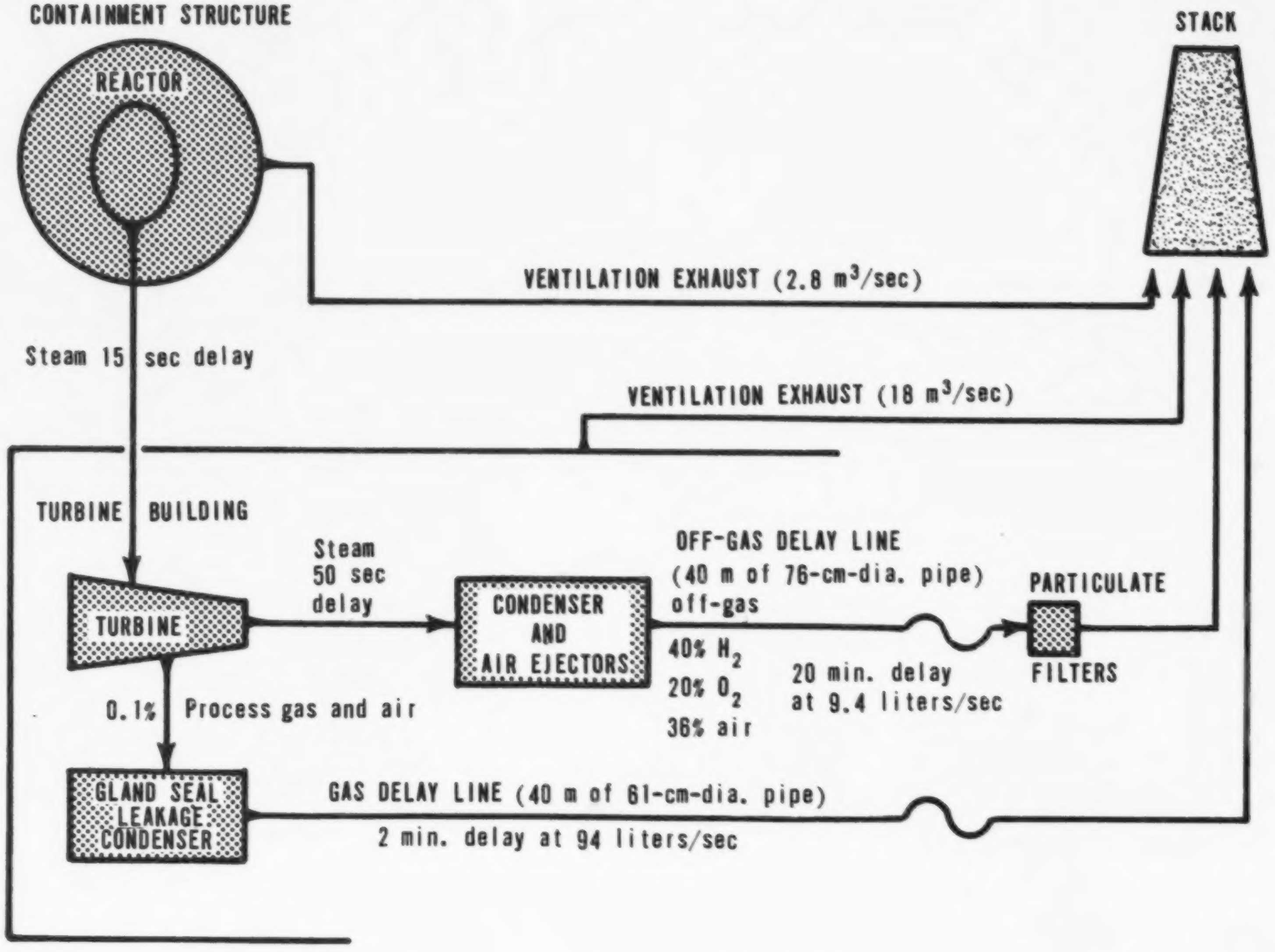
CONTAINMENT STRUCTURE
REACTOR
STACK
VENTILATION EXHAUST (2.8 m³/sec)
Steam 15 sec delay
VENTILATION EXHAUST (18 m³/sec)
TURBINE BUILDING
Steam 50 sec delay
TURBINE
CONDENSER AND AIR EJECTORS
OFF-GAS DELAY LINE (40 m of 76-cm-dia. pipe) off-gas
40% H₂
20% O₂
36% air
20 min. delay at 9.4 liters/sec
PARTICULATE FILTERS
0.1% Process gas and air
GLAND SEAL LEAKAGE CONDENSER
GAS DELAY LINE (40 m of 61-cm-dia. pipe)
2 min. delay at 94 liters/sec

Figure 4.1. Sources of airborne effluent.

body of any individual in an unrestricted area of 0.5 rem in a calendar year, 0.1 rem in seven consecutive days, or 0.002 rem in any one hour. Release limits of individual radionuclides are defined in paragraph 20.106, and concentrations in air are tabulated in Appendix B, Table II, Column 1 of 10 CFR 20 (see Section 3.1.3).

The average activity release rate at the stack to the atmosphere for 1968 while the plant was operating was approximately 12,500 μCi/sec.(5) The reactor was in operation during 64 percent of the year.(5) The gaseous fission product release rate is determined daily at Dresden by collecting a sample of air-ejector off-gas at the delay-line port at 6 A.M., counting it with a single-channel NaI(Tl) gamma-ray detector, and converting count rate to release rate (in μCi/sec) by means of a chart prepared by General Electric Company personnel.(3) The noble gas composition typical of the Dresden stack discharge is listed in Appendix B.3. These data are derived from measurements of radioactive gases at Dresden by General Electric Company personnel,(2) and calculations for a ''diffusion mixture''(6) (see Section 2.3.4).

4.1.2 *Sample collection.* Samples of the off-gas and stack releases were obtained to demonstrate sampling and analytical methods and to determine the composition of effluent from the stack. Emission data were used to estimate radionuclide concentrations in environmental ground-level air and deposition on vegetation. The data also provide comparison with the nominal stack releases reported by Dresden, information on release rates of ^{3}H and ^{85}Kr, and values of collection efficiency for the radioiodine sample.

The off-gas was sampled at the port, located in the delay line 4 minutes downstream from the air ejectors, that is used for daily gaseous release measurements by Dresden staff. Measured concentrations were converted to release rates at the stack for a 16-min transit time from port to stack for radioactive decay, and a flow rate of 9.4 liter/sec.

Off-gas samples were obtained from Dresden staff 9 times in 9-cc glass serum bottles for gamma-ray spectrometry, and twice in 209-cc volumetric flasks for ^{3}H and ^{85}Kr analyses. The bottles and flasks were sealed with rubber stoppers held by crimped aluminum seals. The rubber stoppers permitted insertion of the hypodermic needle outlet of the sampling port. The needle is connected also by means of a two-way valve to a pump for evacuating the container before sampling. The internal pressure of the sample was approximately atmospheric. In tests of container integrity for gas retention with ^{85}Kr or ^{133}Xe for periods as long as 4 weeks, losses were less than 5 percent.

On January 17, 1968, a 209-cc sample of gas was obtained from the stack monitoring system at the same time that a 9-cc gas sample was taken at the off-gas sampling port. The two sets of measured radionuclide concentrations were used to compare release rates.

Release rates of airborne particles in the stack were determined on 17 occasions with the membrane filters that are routinely inserted in the stack monitoring system for 24-hour periods beginning at 2200 or 2300 hours. These samples were dissolved on 5-cm-dia. stainless steel planchets, analyzed as usual by the Dresden staff, and then made available seven days after collection.

Releases of gaseous ^{131}I were measured on the same 17 occasions with charcoal cartridges exposed behind the membrane filters for the same duration. On two occasions, the ^{131}I collection efficiency of the charcoal cartridges was tested by inserting a second cartridge identical to the first in the stack sampling system, followed by two 69-g beds (5.7 cm depth) of MSA Type 85851 charcoal. The latter was impregnated with KI and had been recommended for sampling CH_3I and other organic iodine vapors. Under conditions that are far more rigorous than in the Dresden stack, a 5-cm bed had been found to retain 99% of CH_3I at 140°C and 87% relative humidity.(7)

Because the laboratory was so distant from Dresden that analyses could not be begun until 6 hours after sampling, the set of samples collected on January 17, 1968 was taken to Argonne National Laboratory.* Gamma spectrometric analysis of these samples began 2 hours after sampling. On June 26 and 27, 1968, the off-gas was analyzed within 4 minutes of sampling with a gamma spectrometer in a panel truck parked near Dresden.

Air filters from monitors of ambient air in the containment and turbine buildings were obtained from Dresden staff to assess the contribution of the ventilation air to stack radioactivity. At Dresden, radioactivity of airborne particles is routinely monitored within the containment sphere by means of a device consisting of a detector, a continuously moving strip filter, and an 85-l/min pump. The device is located in front of the inlet of the exhaust duct. In the turbine building, a Schmidt constant flow pump that draws air through a 5-cm-dia. filter at 42 l/min is located at the high pressure end of the turbine. These samples do not accurately represent concentrations of airborne particles in exhaust ducts, but may indicate their magnitude.

4.2 Analysis

4.2.1 *Gamma-ray spectrometry.* Radionuclides that emit gamma rays were analyzed in the laboratory with a 10 × 10 cm cylindrical NaI(Tl) detector coupled to a 400-channel spectrometer (see Figure 4.2). Gamma-ray spectra obtained with a high-resolution 6 cm^2 × 7 mm or 10.4 cm^2 × 11 mm Ge(Li) detector and a 1600-channel spectrometer assisted in identifying radionuclides (Figure 4.3). Samples taken to Argonne National Laboratory were analyzed by single and dual 10 × 10 cm NaI(Tl) crystals and a small Ge(Li) detector (see Figure 4.4). The counting system carried by truck to Dresden consisted of a 10 × 10 cm NaI(Tl) detector mounted in a lead-brick shield, a 400-channel spectrometer (see Figures 4.5 and 4.6 for typical spectra), a gasoline-fueled alternator to provide power, and a Sola transformer.

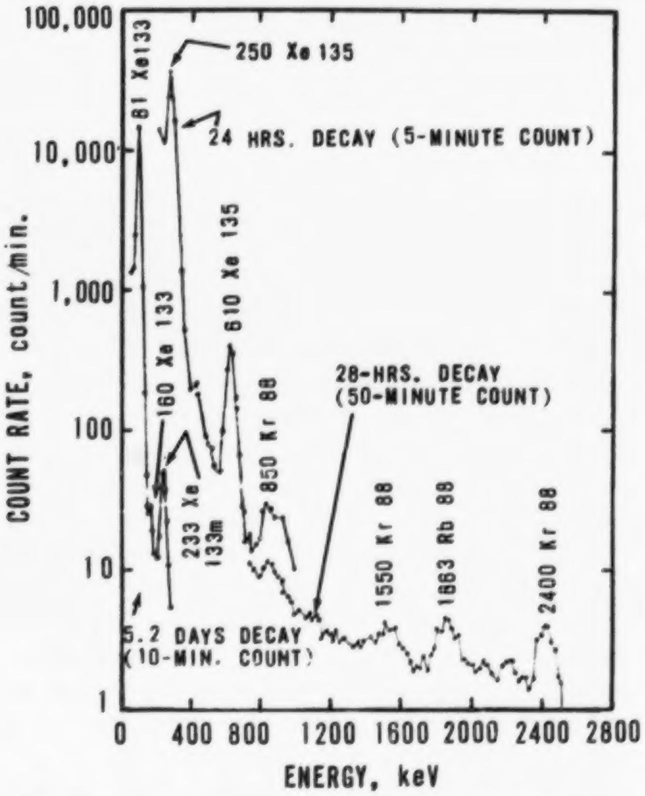

Figure 4.2. Gamma-ray spectra of off-gas from delay line. Detector: NaI(Tl), 10 × 10 cm. Sample: 9-cc bottle of gas collected 0900 hour EST, Nov. 16, 1967.

Samples were analyzed in their original containers by gamma-ray spectrometry. When possible, detection efficiencies for the containers and radionuclides of interest were determined with standardized radioactivity solutions. Accurate analysis of gases was hindered by the unavailability of standardized radioactive gases; for some radionuclides, even pure unstandardized isotopes that would provide individual spectra for calibration were not available. Hence, calibration curves of counting efficiencies *vs.* photon energy were established by normalizing the shape of efficiency curves for other samples to the efficiency value of the ^{85}Kr 514-keV gamma ray. The uncertainty of this curve is estimated to be 10 percent or less.

The radioactivity in the Cesco cartridge was concentrated on the upstream portion, hence measurements were made on both sides and this distribution was simulated in the calibration standard. Analysis was delayed

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*We thank Dr. Jacob Sedlet and his associates at Argonne National Laboratory for analyzing these samples.

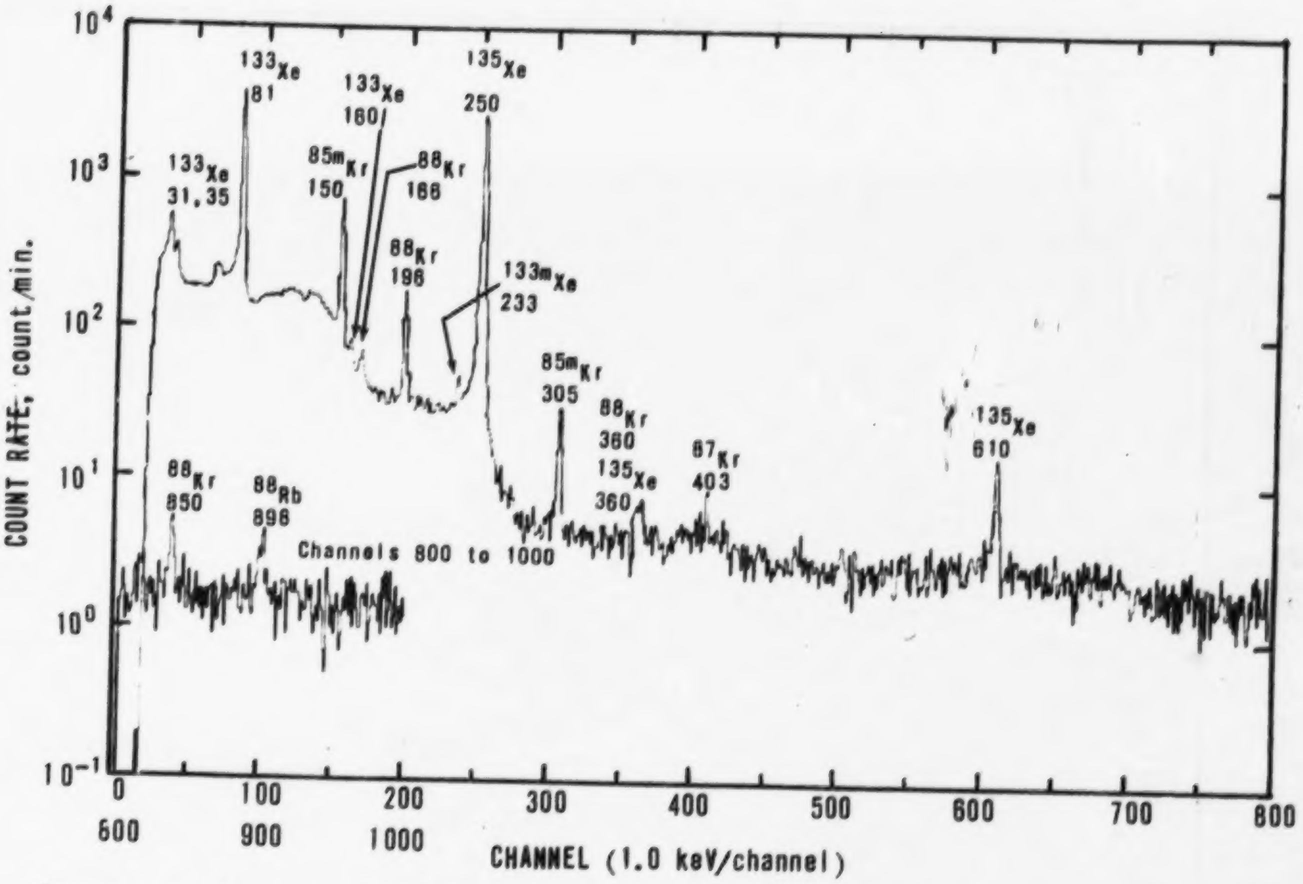

Figure 4.3. Gamma-ray spectrum of off-gas from delay line. Detector: Ge(Li), 6 cm^2 × 7 mm. Sample: 9-cc bottle of off-gas collected 1030 hour EST, Jan. 18, 1968. Count: Jan. 18, 2130 – 2140 hour.

several days to await reduction by radioactive decay of radio-xenon (particularly ^{135}Xe) and progeny that initially obscured the ^{131}I peak. Because of the delay, no short-lived radioiodine was detected, and only gamma rays from ^{131}I, ^{133}Xe, and the 30-yr ^{137}Cs daughter of ^{137}Xe were observed.

Counting intervals and techniques were selected to provide a counting error of ± 5% or less at the 95 percent (2-sigma) confidence level for most radionuclides. Counting of low-level radioactivity was usually for 1000 min. Samples were reanalyzed periodically to check the half lives of identified radionuclides and to measure longer-lived radionuclides that had previously been obscured by radiations from shorter-lived radionuclides, as indicated in Figures 4.5 and 4.6.

Complex sample spectra were analyzed by comparison with the spectral shapes of the individual components. Because separated short-lived radioactive gases were unobtainable for spectral calibrations, analytical uncertainties for these radionuclides may be as large as a factor of two.

4.2.2 *Radiochemical analysis.* After gamma spectral analysis was completed, strontium on the membrane filters and ventilating-air samples was chemically separated and measured for ^{89}Sr and ^{90}Sr content with low-background G-M beta counters at 100-min. counting periods.

Tritium and ^{85}Kr concentrations in the 209-cc gas samples were determined by liquid scintillation counting after the decay of shorter-lived radionuclides. To collect ^{3}H in water from either gas (HT) or water vapor (HTO), 20 ml of water were injected into the 209-cc sample containers and occasionally swirled for 3 to 8 days. The water was then transferred and distilled, and aliquots were mixed with liquid scintil-

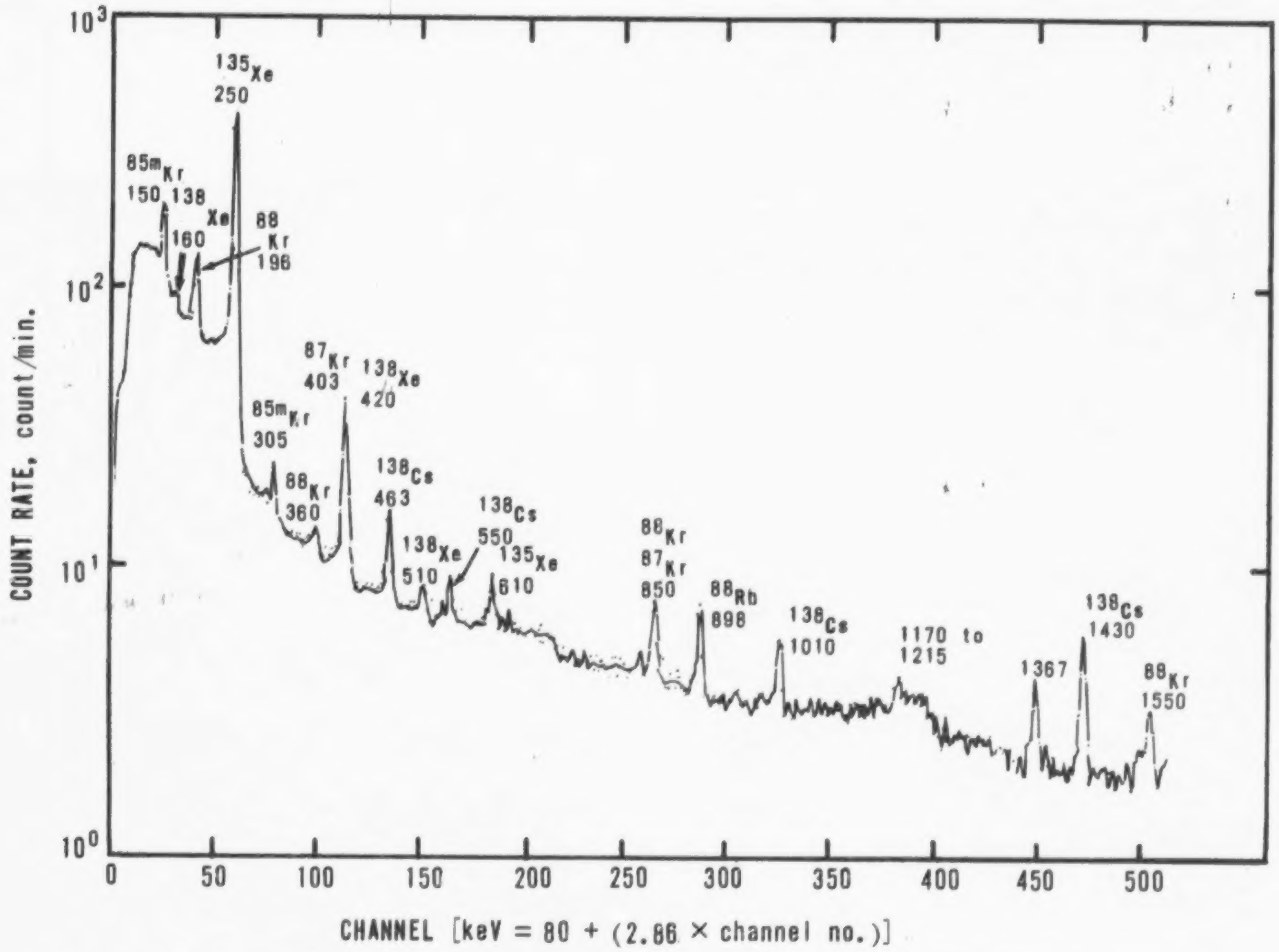

Figure 4.4. Gamma-ray spectrum of off-gas from delay line. Detector: Ge(Li). Sample: 9-cc bottle of off-gas collected 1100 hour EST, Jan. 17, 1968. Count: Jan. 17, 1407 – 1417 at ANL.

lator for analysis.* The difference between the count rates in the water samples and in water blanks was attributed to tritium from the gas samples. The resulting net tritium concentration was multiplied by 189/20 to calculate the tritium concentration in off-gas. Gas for ^{85}Kr analysis* was withdrawn from the sample bottles when the water was introduced, mixed directly with degassed liquid scintillator, and analyzed.$^{(8)}$

4.3 Results and Discussion

4.3.1 *Radioactive noble gases and ^{3}H.* The prominent radionuclides in stack effluent, as measured by gamma spectral analysis of delay-line samples, were ^{85m}Kr, ^{87}Kr, ^{88}Kr, ^{133}Xe, ^{135}Xe, and ^{138}Xe (see Table 4.1). Xenon-135m is the only radioactive noble gas among the major predicted constituents of the stack effluent (Appendix B.3) that could not be quantified. One day after release, only ^{88}Kr, ^{133m}Xe, ^{133}Xe and ^{135}Xe were detected by gamma-ray spectrometry (see Figure 4.2); after one month, only ^{133}Xe, ^{85}Kr and ^{3}H remained. The rapid initial decrease in radioactivity is indicated in Figures 4.5 and 4.6.

The release rates of the individual radioactive noble gases and their amounts relative to nominal gaseous fission product values in Table 4.1 vary appreciably among samples. As indicated by the data summary in Table 4.2, releases of individual radionuclides, normalized to 1 mCi of nominal gaseous fission product release, ranged

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*We thank Dr. A. Moghissi and staff at the Southeastern Radiological Health Laboratory for analyzing these samples.

Table 4.1

Stack Releases of Fission Product Noble Gases*, μCi/sec

Radionuclide	Sample Date and Time (Central Time)								
	11/15/67 (0600)	11/16/67 (0600)	1/17/68 (1000)	1/18/68 (0930)	1/31/68 (0900)	6/26/68 (1030)	6/27/68 (1630)	8/20/68 (1928)	8/21/68 (0547)
4.4 -hr ^{85m}Kr	---	580	340	280	---	---	---	---	---
10.7 -yr ^{85}Kr	---	---	---	---	---	0.024	---	---	0.25
76 -m ^{87}Kr	---	---	~540	~540	---	~1,300	---	---	---
2.8 -hr ^{88}Kr	---	~1,300	~780	~640	---	~260	---	---	---
2.3 -d ^{133m}Xe	---	14	11	11	---	8	5	31	20
5.3 -d ^{133}Xe	450	420	400	410	970	160	110	910	730
9.1 -hr ^{135}Xe	930	1,010	1,660	1,250	2,600	620	520	---	1,420
17 -m ^{138}Xe	---	---	~850	---	---	~2,800	~2,400	---	---
Gaseous fission product release rate	11,000	10,000	14,800	12,600	18,400	7,500	5,700	11,600	11,600

Based on sampling off-gas delay line; computed for release at top of stack after radioactive decay and dilution of 9.4 l/sec off-gas by exhaust air from containment and turbine buildings at flow rate of 21 m^3/sec.

Blank space indicates that radionuclide was not measured.

Table 4.2

Observed vs. Estimated Stack Release Rates of Noble Gases

Radionuclide	Number of measurements	Observed rate*		Estimated rate*	Observed mean / Estimated
		Range	Mean		
^{85m}Kr	3	22–58	34	32	1.06
^{85}Kr	2	0.003–0.022	0.012	0.052	0.23
^{87}Kr	3	37–180	~85	102	~0.83
^{88}Kr	4	35–130	~67	109	~0.62
^{133}Xe	9	19–79	42	33	1.26
^{135}Xe	8	83–140	104	114	0.91
^{138}Xe	3	57–420	~284	243	~1.17

*Normalized as μCi of radionuclide per mCi of nominal gaseous fission product release.

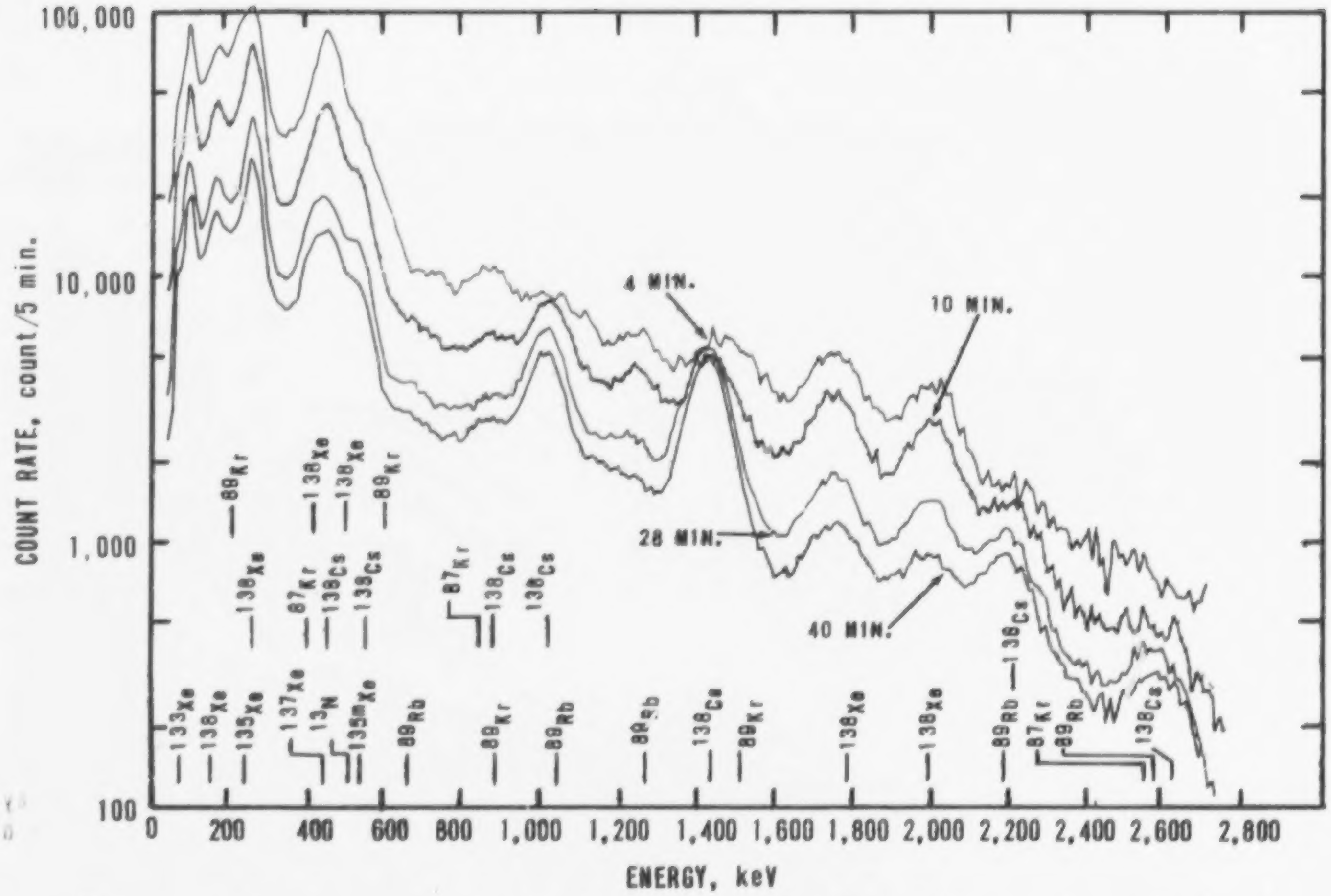

Figure 4.5. Gamma-ray spectra of off-gas from delay line after various periods of decay. Detector: 10 × 10 cm NaI(Tl). Sample: 9-cc bottle of gas collected 1613 hrs. CDT, June 27, 1968. Count: Sample was centered 24.3 cm above detector. Times shown are from sampling to midpoint of counting interval.

within approximately a factor of two of their mean values. The ratio of the mean measured value to that estimated in Appendix B.3 for a 21-minute delay period ranged from 0.6 to 1.1 for all gases except ^{85}Kr. The measured release rate of ^{85}Kr would be expected to be lower than in a mixture that contained both equilibrium and recoil components (see Section 2.3.4) because the period of reactor operation was too short for ^{85}Kr transfer to reach equilibrium. The agreement of most measured and predicted release rates within a factor of two confirms the applicability of the constant determined by Gilbert(2) for equation 2.3.

In the single comparison of radioactive noble gases in the stack and in the delay line (Table 4.3), values in the stack were consistently higher. The differences may be due to incomplete mixing of off-gas with ventilating air below the stack monitor, calculation of release rates with incorrect flow rates in delay line, stack or stack sampler, error in standardizing the gamma-ray spectrometer for the gas containers, or sources of radioactive noble gases in addition to delay-line off-gas. More measurements are needed to resolve the differences.

The gamma-ray spectra of off-gas measured within a few minutes after collection (see Figures 4.5 and 4.6) indicated the presence of 10-min ^{13}N, which is formed by the reaction $^{16}O(p,\alpha)^{13}N$ and is reported to be a major constituent of the radioactive stack effluent.(2) Its release rate could not be quantified, however, because of spectral interference by other short-lived radionuclides, especially ^{135m}Xe.

Tritium concentrations in the off-gas and tritium release rates computed from

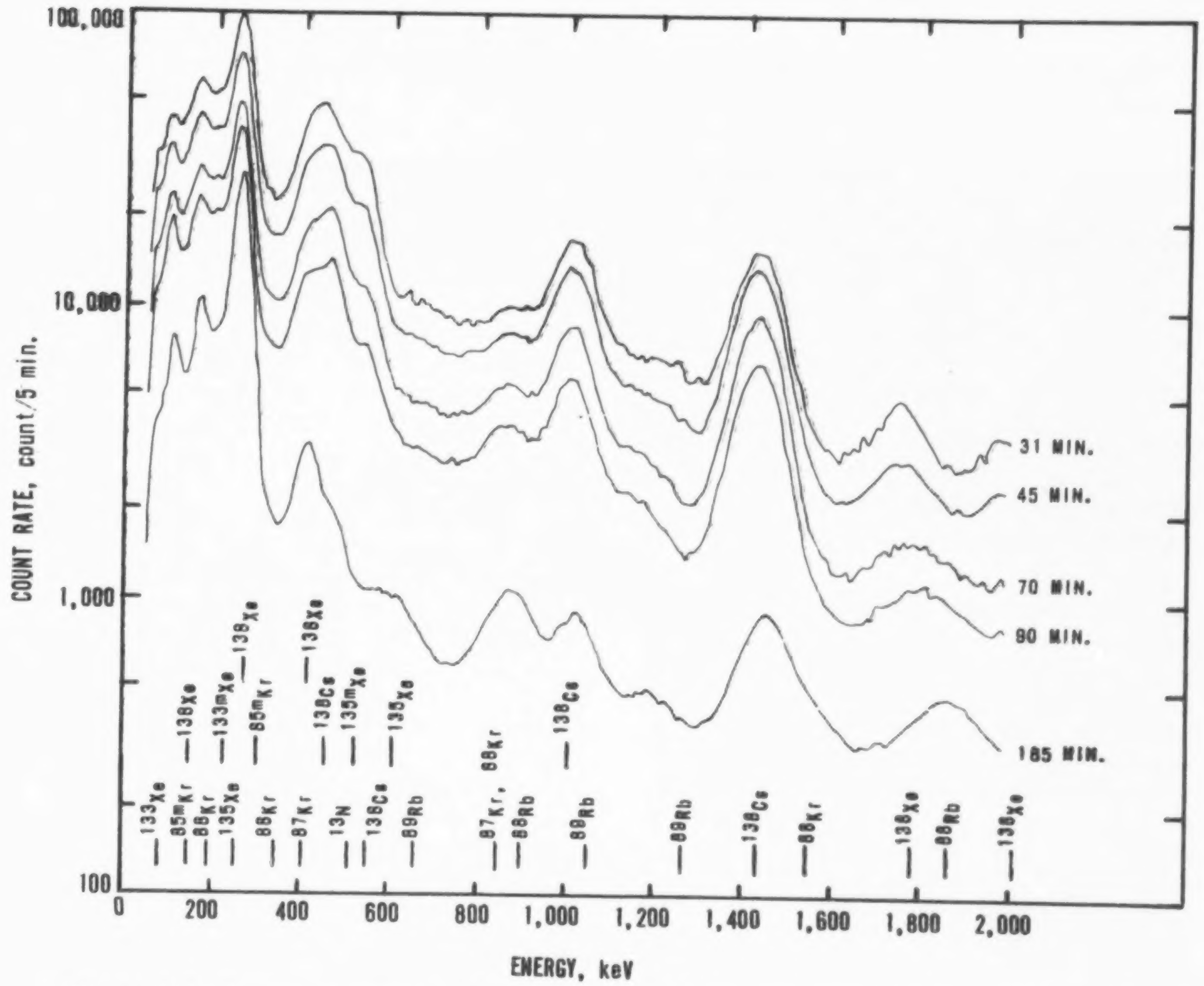

Figure 4.6. Gamma-ray spectra of off-gas from delay line after various periods of decay. Detector: 10 × 10 cm NaI(Tl). Sample: 9-cc bottle of gas collected 1030 hrs. CDT, June 26, 1968. Count: Sample centered 9.8 cm above detector. (Spectrum of 185-min. count was adjusted to simulate counting at this distance). Times show interval from sampling to midpoint of counting.

these concentrations are given in Table 4.4. Also shown are release rates estimated from the tritium concentration in primary coolant on the assumptions that the off-gas is 40 percent hydrogen from the dissociation of water in the reactor,(1) at a temperature of 32°C (at which the volume of 1 mole of H_2 is 25 liters), and at 100 percent humidity. Thus, the amount of water from which the hydrogen in the off-gas was derived is

$$\frac{9.4 \text{ liter/sec} \times 0.40 \times 18 \text{ ml } H_2O\text{/mole}}{25 \text{ liter } H_2O\text{/mole}}$$

$$= 2.7 \text{ ml } H_2O\text{/sec}$$

Water vapor contributes another 34×10^{-3} ml H_2O/liter of gas × 9.4 liter of gas/sec = 0.32 ml/sec, for a total release of primary-coolant water of 3.0 ml/sec in off-gas. Release rates measured on June 26 agreed

Table 4.3

Comparison of Off-gas Release Rates Measured in Delay Line and in Stack, μCi/sec

Radionuclide	Sampling Point: Delay Line*	Sampling Point: Stack	Stack/Delay Line
^{85m}Kr	340	520	1.5
^{87}Kr	~540	~1240	2.3
^{88}Kr	~780	~910	1.2
^{133}Xe	400	650	1.6
^{135}Xe	1660	1970	1.2

*Computed for release at top of stack after radioactive decay and dilution of 9.4-l/sec off-gas by exhaust air from containment and turbine buildings at flow rate of 21 m^3/sec.

with the predicted values but the value for January 18 was 3-fold higher. Measured and estimated tritium concentrations may differ because the primary coolant water and the off-gas were not sampled at the same times, or because of erroneous assumptions concerning the content of hydrogen and water vapor in off-gas. The analytical procedure depends on complete exchange of tritium in HT with the water added to the sample, an assumption that should be tested in view of reported slow exchange rates.(9) In future studies, the total tritium release rate in stack gas should also be measured.

4.3.2 *Airborne particles in the stack.* The longer-lived particulate radionuclides observed in the stack were the activation products ^{58}Co and ^{60}Co, and the gaseous-fission-product progeny ^{89}Sr, ^{90}Sr, ^{137}Cs and ^{140}Ba. Emission rates measured for 17 samples are shown in Table 4.5, and mean values and ranges, in Table 4.6. Activity levels and relative composition fluctuated considerably, and no relation was found between release rates of particles and nominal gaseous fission products. Particulate ^{131}I was never found above the minimum detectable emission rate of 30 pCi/sec, compared to release rates of gaseous ^{131}I between 200 and 3230 pCi/sec.

4.3.3 *Gaseous ^{131}I in the stack.* The gaseous ^{131}I release rates observed in the stack with the routinely used single charcoal cartridge are given in Tables 4.5 and 4.6. The test of cartridge collection efficiency indicated that 88 percent of the collected ^{131}I is retained on the usual sampling cartridge and the remaining 12 percent is on a second cartridge (Table 4.7). These values agree with a similar test conducted with filtered gas from the CP-5 reactor, wherein 95 percent was retained by a single Cesco cartridge and 98.7 percent by two in series.(10) Dresden staff reports that the conservative factor for calculating ^{131}I release rates from count rates more than compensates for incomplete radioiodine retention in the cartridge.(3)

No ^{131}I (< 1.2 percent) was found on the KI-impregnated charcoal beds located behind the Cesco cartridges. This type of charcoal is reported to be an effective collector

Table 4.4

Release Rate of Tritium from Condenser Air Ejectors

Date, 1968	Concentration in primary coolant, pCi/ml	Concentration in delay line, pCi/cc	Release rate, nCi/sec: Measured	Release rate, nCi/sec: Estimated*
Jan. 18		1.5	14	---
Feb. 1	1,660		---	5.0
June 26		0.52 ± 0.03†	4.8 ± 0.3	---
Aug. 22	1,300		---	3.9

*based on equivalent water for hydrogen gas of 2.7 ml/sec and for water vapor of 0.32 ml/sec.

†standard deviation of duplicate analyses; only one sample was analyzed on Jan. 18.

Note: 1 nCi/sec = 1 x 10^{-3} μCi/sec.

Table 4.5

Stack Releases of Particulate Radionuclides and Gaseous Iodine-131, pCi/sec

	Sampling Period (~24 hours)															
	Nov. 1967		Jan. 1968			June 1968									Aug. 1968	
Radionuclide	7	8	9	10	17	19	20	21	22	23	24	25	26	27	28	20	21
Particles on membrane filter																
71 -d ^{58}Co	< 8	NA	NA	17	140	< 8	29	13	< 8	< 8	< 8	9	< 8	< 8	< 8	70	120
5.26-yr ^{60}Co	27	21	20	NA	20	25	23	2.5	8	10	19	18	11	23	12	60	95
51 -d ^{89}Sr	NA	420	2300	1170	NA	1280	860	1180	1310	800	920	580	900	850	1180	610	220
28 -yr ^{90}Sr	NA	5	6	25	NA	3	2	3	3	2	3	3	3	2	3	2	3
30 -yr ^{137}Cs	25	NA	48	50	13	55	38	41	36	29	31	31	34	38	38	30	25
12.8 -d ^{140}Ba	360	250	480	390	70	480	500	510	710	580	700	400	500	440	400	440	85
Gaseous ^{131}I on charcoal cartridge																
8.06-d ^{131}I	360	330	3230	2560	2780	250	930	870	200	240	220	820	1060	600	870	840	210
Gaseous fission product release rate, (mCi/sec)	8.7	17.0	18	53	15	7.4	6.6	7.1	5.9	5.8	5.7	6.4	7.5	5.7	6.7	11.6	11.6

NA – No analysis made.

< values indicate minimum detectable activity at the ~3 σ confidence level.

1 pCi/sec = 1 x 10^{-6} μCi/sec.

1 mCi/sec = 1,000 μCi/sec.

Table 4.6

Summary of Stack Releases of Particulate Radionuclides and Gaseous Iodine-131, pCi/sec

Radionuclide	Number of Measurements	Mean	Range
^{58}Co	15	26*	< 8 - 140
^{60}Co	16	25	2.5 - 95
^{89}Sr	15	970	220 - 2300
^{90}Sr	15	5	2 - 25
^{137}Cs	16	35	13 - 55
^{140}Ba	17	430	70 - 710
^{131}I	17	920	200 - 3230
Gaseous fission product release rate, mCi/sec	17	11.7	5.7 - 53

*values below minimum detectable activity values were taken to be zero for averaging.

1 pCi/sec = 1 x 10^{-6} μCi/sec

of CH_3I under conditions that lead to penetration of Cesco-type charcoal by CH_3I.(7) Thus, essentially all of the ^{131}I released at the stack appears to be in a form that can be collected on the Cesco cartridges.

4.3.4 *Airborne particles in ventilating air.* Radionuclide concentrations in air, measured on filters that were collected in the turbine and containment buildings on one occasion are listed in Table 4.8. Radionuclide release rates in ventilating air were computed from ventilation rates on the assumption that the sampled concentrations were representative of values in the exhaust duct. Compared to stack-monitor values on January 16–17 (Table 4.5), release rates in ventilating air were of the same magnitudes for ^{90}Sr and ^{137}Cs, but much smaller for ^{58}Co, ^{60}Co, ^{89}Sr, ^{131}I, and ^{140}Ba. Note that ^{131}I was collected on filters (i.e., as particles) in the ventilating air and on charcoal (as gas) in the stack effluent.

Table 4.7

Iodine-131 Concentration in Stack Monitor Exposed 2120 CDT August 20 to 2125 CDT August 21, 1968

Sampler	^{131}I, pCi/m^3
First Cesco cartridge	10 ± 0.5
Second Cesco cartridge	1.3 ± 0.2
KI-impregnated charcoal	<0.15

< value indicates minimum detectable activity at the ~3 σ confidence level.

4.3.5 *Sources of airborne particles.* The airborne particulate radionuclides detected in the stack sampler may reach the stack (1) through ingrowth from gaseous precursors between the high efficiency particulate air filter in the air-ejector off-gas line and the stack monitor, (2) by penetrating the filter, (3) in off-gas from the turbine gland seal condenser and (4) in ventilation air from turbine building and containment shell.

Contribution by sources (1), (2) and (3) can be estimated from Gilbert's values(2) for radioactive gases at the air ejector per mCi/sec of nominal gaseous fission product discharge. These contributions were estimated for 4 radionuclides from the stack measurements from June 18 to 28, 1968, when the noble gas discharge averaged 6.5 mCi/sec. The values given below are based on decay and ingrowth calculations for a 20-minute delay from the air-ejectors to the filter, 0.1 percent filter penetration, and a 10-second delay from the filter to the stack monitors. Also assumed were a 2-minute delay for gland seal condenser off-gas, and a 0.1 percent leakage of process gas through the gland seals.(1)

			Computed release rate at stack sampler*, pCi/sec				
Radionuclide	Precursor gas at air ejector, mCi/sec		Ingrowth after filter	0.1% filter leakage	Gland seal off-gas	Total	Measured mean, pCi/sec
^{89}Sr	5.7	^{89}Kr	140	240	100	480	990
^{90}Sr	4.9	^{90}Kr	< 0.1	0.2	< 0.1	0.2	2.6
^{137}Cs	6.9	^{137}Xe	1.3	1.6	1.8	5	37
^{140}Ba	1.2	^{140}Xe	< 0.1	17	150	167	520

*Includes production by ^{89}Rb and ^{140}Cs, parents of ^{89}Sr and ^{140}Ba, respectively, after collection on the sampler.

Table 4.8

Airborne Particles in Sphere and Turbine Building

Radionuclide	Concentration, pCi/m^3		Estimated release rate, pCi/sec			Release rate in stack, pCi/sec
	Sphere	Turbine	Sphere	Turbine	Total	
^{58}Co	2.1	1.0	5.9	18	24	140
^{60}Co	1.2	< 0.1	3.4	< 1.8	~3.4	20
^{89}Sr	3.2	3.0	9.0	54*	63	2,300
^{90}Sr	0.11	0.35	0.3	6.3	6.6	6
^{131}I	16	< 0.5	45	< 9.0	~45	2,780
^{134}Cs	0.4	< 0.1	1.1	< 1.8	~1.1	---
^{137}Cs	1.2	0.9	3.4	16.2	20	13
^{140}Ba	0.4	0.9	1.1	16.2	17	70

Notes: 1. Ventilation exhaust rates to the stack are 2.8 m^3/sec from the containment sphere and 18 m^3/sec from the turbine building.

2. < values indicate minimum detectable activity at the 3 σ confidence level.

3. Sampling intervals: containment sphere - 1445 hrs., Jan. 17, 1968 to 0900, Jan. 18.
turbine - 0000 to 2400 hrs., Jan. 17.
stack - 2250, Jan. 16 to 0910, Jan. 17, then 1100 to 2300, Jan. 17, except that ^{89}Sr and ^{90}Sr rates are for Jan. 9, 1968.

Estimated releases of ^{89}Sr and ^{140}Ba are lower than measured rates by factors of 2 and 3, respectively. Higher values would result from larger-than-assumed amounts of precursor gases, higher filter or gland seal leakage, or appreciable contributions from ventilating air. The much lower estimates of ^{90}Sr and ^{137}Cs contributions compared to measured rates support the finding that ventilating air exhaust is the principal source of ^{90}Sr and ^{137}Cs in the stack monitor.

4.4 References

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5. RADIONUCLIDES IN ENVIRONMENTAL AIR

5.1 Introduction

5.1.1 *Purpose.* These studies were undertaken to demonstrate sensitive and convenient techniques for detecting the plume released at BWR stacks, measuring radionuclide concentrations in the plume, and determining the radiation exposure rate associated with the plume. At BWR stations, the critical pathway appears to be from the stack through air, leading to whole-body irradiation of persons in or beneath the plume at ground level near the station. By comparing, under appropriate conditions and with sufficient frequency, radionuclide concentrations in the environment with release rates at the stack, the degree of atmospheric diffusion for computing release limits is determined; by comparing exposure rates in the environment with release rates at the stack, release limits are determined directly.

Survey meters were used to detect the plume. For brief periods, (1/2 to 1 hour) the radiation exposure rate was measured with sensitive muscle-equivalent ionization chambers, radioxenon was collected with a cryogenic apparatus developed for this purpose, and a radioactive particulate daughter of radioxenon was collected with high-volume air samplers. For longer periods (2 weeks), radiation exposure was measured with thermoluminescent dosimeters (TLD's). Both methods of measuring radiation exposure were used because there is considerable uncertainty in computing long-term exposure from brief observations and also in measuring long-term exposure near background values.

5.1.2 *Environment and meteorology.* The environment of Dresden is sparsely wooded and flat, except for a bluff 30 m high on the far bank of the Illinois River, 1 km north and east of the stack. (For further description of the environment, see Section 7.1.1). Winds blow predominantly from the southwest during most of the year,(1) but westerly winds are frequent during the winter. Rainfall is heaviest in summer. Summer rains are from convective showers and thunderstorms, winter rains from passing cyclones.

Meteorological data for planning and interpreting the experiments were obtained from an instrumented 123-m tower located 0.8 km SSW of the plant. Wind speed and direction instruments and temperature sensors are in the tower at levels 11, 38, 92, and 123 m above ground. Data are recorded continuously on strip charts and were provided on request by Dresden staff. The data are also summarized by computer for wind speed, direction, and direction variance for one 15-minute average per hour.*

5.2 Short-term Radionuclide Concentrations and Radiation Exposure Rates

5.2.1 *Measurements.* Three field experiments were conducted on the bluff north to east of Dresden while the plume from the stack was over the measurement locations. The sites, shown in Figure 5.1, are between 1.6 and 1.8 km distant from the stack. They were selected by observing current wind directions at the Dresden meteorological

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*We thank Dr. M. I. Goldman and Mr. P. Altomare, NUS Inc., for providing this information.

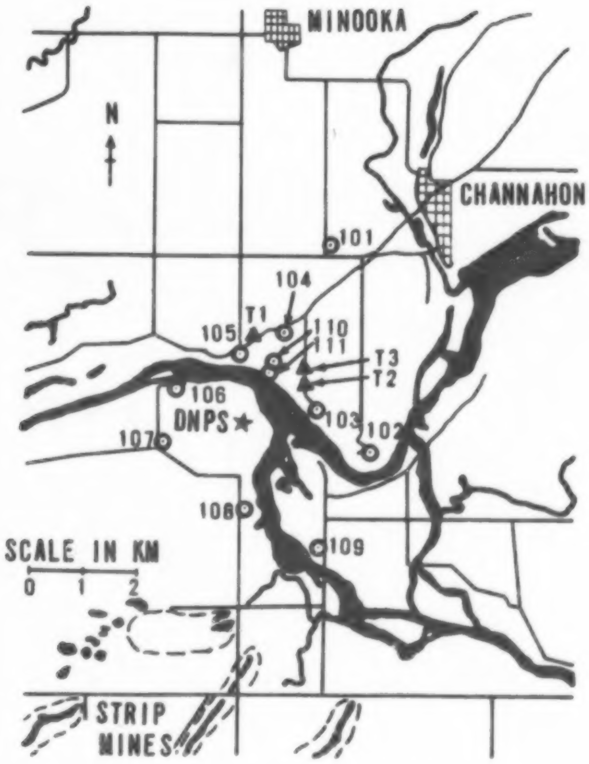

Figure 5.1. Location of field tests (▲) and dosimeter stations (○) near Dresden.

tower, and then traversing roads in the predicted direction with portable NaI(Tl) survey meters (see Section 5.4) to find the center-line of the plume. In several other experiments, wind directions were too variable to permit data averaging over 1/2 to 1 hour periods. Possible sites were limited to existing roads, although the Dresden area is favorable because of the checkerboard pattern of roads at 1-mile intervals. Measurement periods were selected so that the first and third experiments were at neutral conditions during which the instruments were probably within the plume; the second experiment was under stable conditions, with the instruments presumably beneath the plume.

In all three tests, radioxenon was collected 1.5 m above ground by pumping air through charcoal at −80°C (see Section 5.2.2 for details of the apparatus). Volumes of 0.65, 0.68, and 0.40 m^3 were passed through the charcoal in tests 1, 2, and 3, respectively. In the second and third tests, radioactive airborne particles were collected at the same locations by pumping air at the rate of 1.4 m^3/min through a glass fiber filter (MSA CT-75428, 20 x 26 cm) with conventional high-volume air samplers. The charcoal and the filters were analyzed by gamma-ray spectrometry with 10 x 10 cm NaI(Tl) detectors and multichannel analyzers. Two air filters from duplicate systems were counted together each time. The filters represented 157 m^3 of air in test 2 and 96 m^3 in test 3. Spectra obtained immediately after collection during the second and third experiments with detector and analyzer mounted in a light truck that was parked away from the plume are shown in Figures 5.2 and 5.3. All samples were analyzed again in the laboratory, yielding gamma-ray spectra such as the one shown in Figure 5.4. Field spectra were less precise because of shorter counting times and less shielding.

Concurrently with the collection of radionuclide samples, the radiation exposure rate was measured with a muscle-equivalent ionization chamber and Shonka electrometer.(2,3) The ionization chamber is a 16.5-liter sphere with 6-mm-thick plastic walls that contains a neon-ethylene-ethane-nitrogen gas mixture (760 torr at 15°C). The vibrating-reed electrometer has a design sensitivity of 0.32 mV $sec^{-1}/\mu R\ hr^{-1}$, compared to the value measured with a radium standard of 0.33 mV $sec^{-1}/\mu R\ hr^{-1}$ (0.34 mV $sec^{-1}/\mu rad\ hr^{-1}$). At the electrometer operating voltage of 1000 mV, reading periods ranged from 5.5 min for background to 1 min at the highest exposure rates. As many readings as possible were obtained during each experiment. The background exposure rate, measured when the plume passed in another direction, was subtracted from each gross exposure rate. The average exposure rate was computed from these readings, weighted for reading interval.

5.2.2 *Radioxenon collector.* The apparatus was developed to collect quantitatively sufficient radioxenon in a 1-hour period for measurement by gamma-ray spectrometry. It consists of the following components:

(1) flow meter;

(2) 90 g molecular sieve (Linde MS 13 X) in a 4-cm-dia. glass column to remove CO_2 and water vapor;

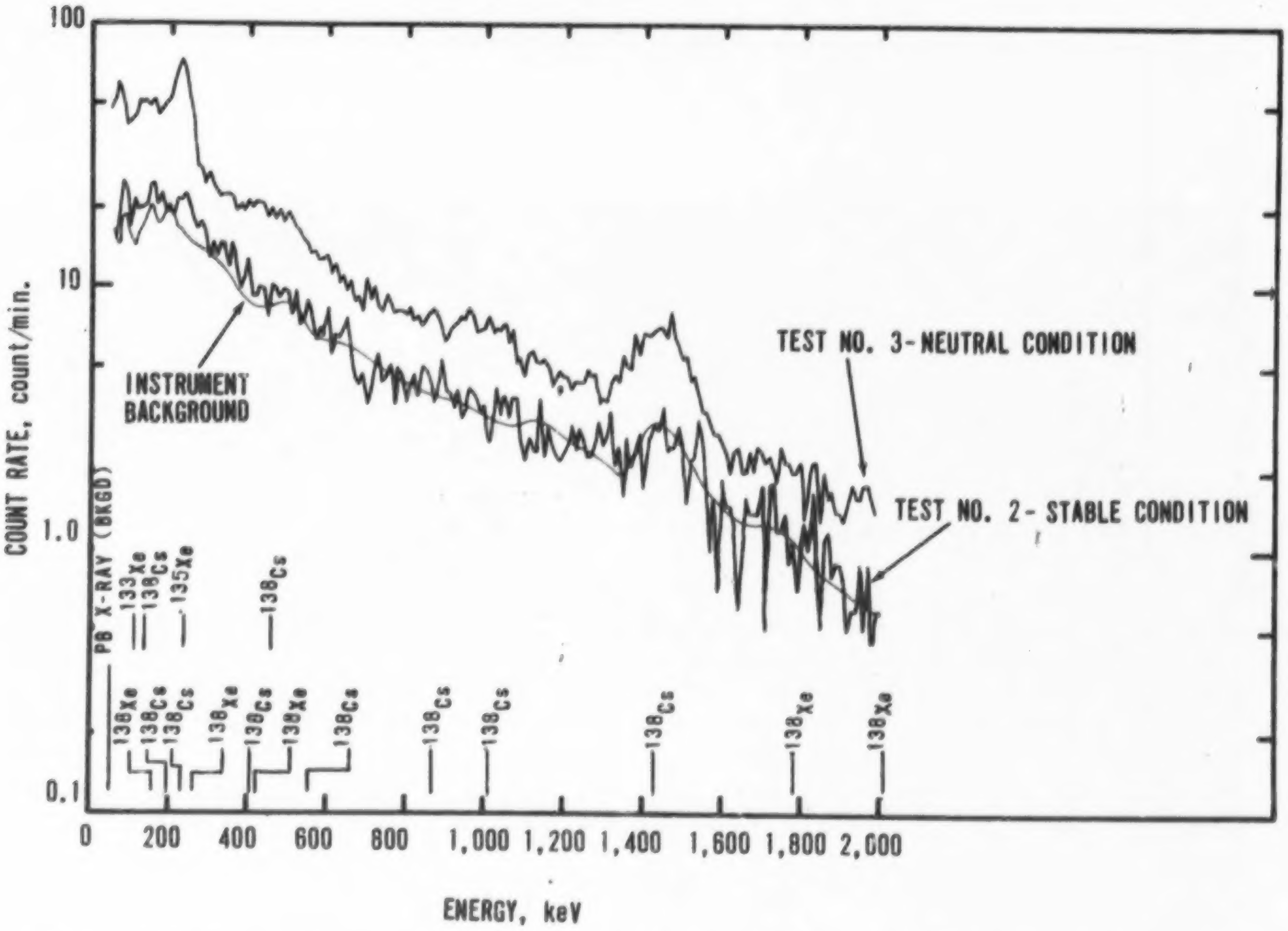

Figure 5.2. Gamma-ray spectra of charcoal xenon collector. Detector: 10 × 10 cm NaI(Tl). Sample: 210 g charcoal in 450-cc gas-tight container, collected Aug. 21, 1968. Counts: Test 2: Aug. 21, 0311–0321; Test 3: Aug. 21, 1905–1925.

(3) copper coil, 1-cm-dia. x 80 cm, for cooling air by immersion in dry-ice-acetone bath;

(4) 210 g charcoal (Columbia 6GC 10/20 mesh) in a 3.2-cm-dia. x 66 cm copper U-tube, to collect radioxenon when immersed in dry-ice-acetone bath;

(5) 30-liter/min vacuum pump;

(6) dry-ice-acetone bath for immersing copper coil and U-tube.

The first five components are connected in the specified order with plastic tubing; the glass column, copper coil and copper U-tube are sealed with 1-hole rubber stoppers and tape. To collect xenon, air is pumped at 15 liter/min for as much as 1 hour. Immediately after pumping, the U-tube is opened, the charcoal is transferred to a plastic 450-cc container, and the container is sealed with a rubber-gasketed, bolted-down lid. The container must withstand pressures of several atmospheres as the charcoal warms up to ambient temperatures. The charcoal in the container is analyzed by gamma-ray spectrometry, either immediately to measure ^{138}Xe, or later, to measure ^{133}Xe and ^{135}Xe.

The apparatus was tested with ^{133}Xe and ^{85}Kr tracers. At the indicated amount of charcoal and flow rate of air, no radioxenon was lost from 1 m^3 of air. Radiokrypton, however, began to break through the column after passage of 0.12 m^3 of air, and can therefore not be collected quantitatively from larger volumes under the defined conditions. These results agree with earlier studies of radiokrypton and radioxenon adsorption on charcoal.(4)

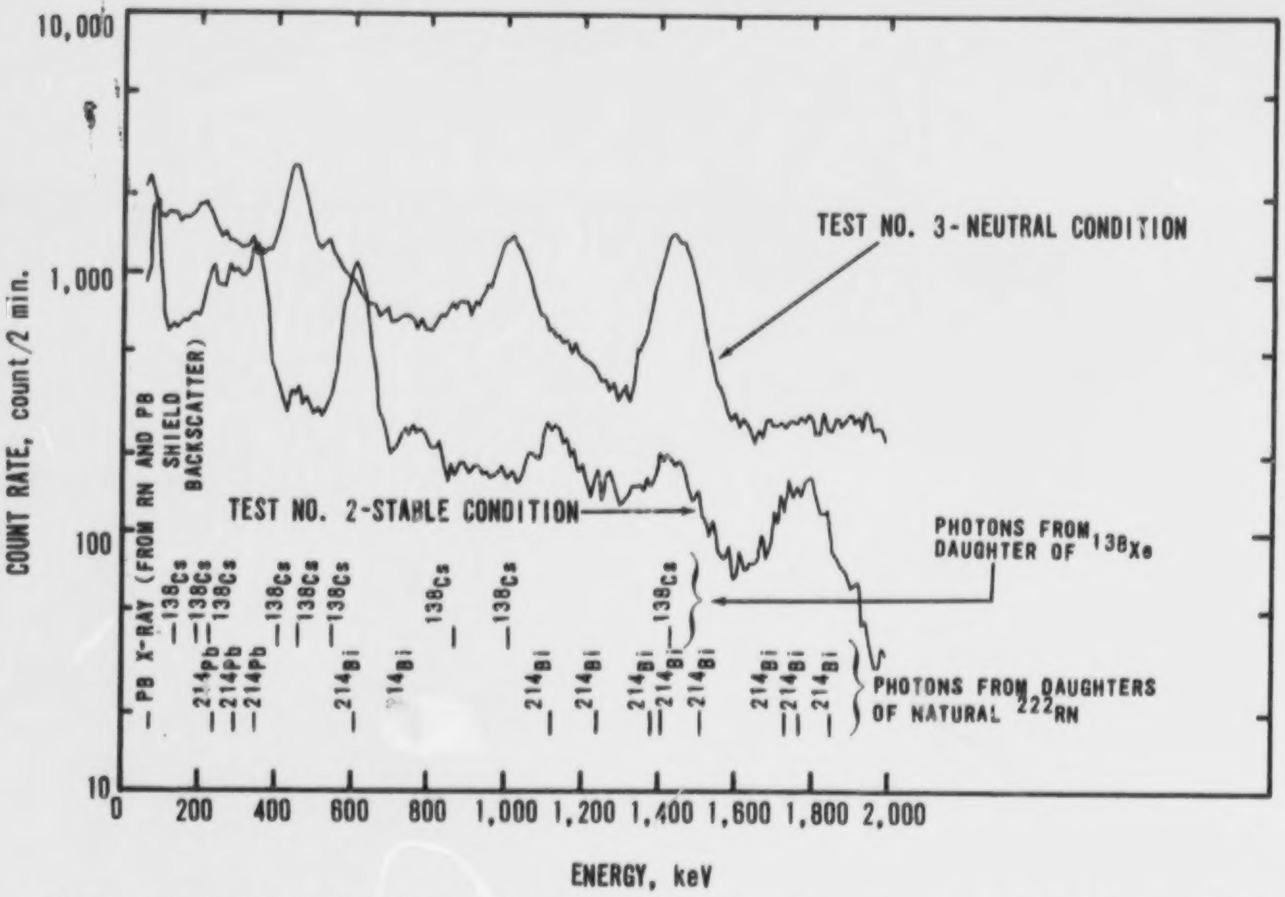

Figure 5.3. Gamma-ray spectra of air filters. Detector: 10 × 10 cm NaI(Tl). Samples: Two 20 × 25 cm glass fiber filters folded into quarters for counting, collected Aug. 21, 1968. Counts: Test 2: Aug. 21, 0325–0327; Test 3: Aug. 21, 1900–1902.

5.2.3 *Meteorology*. The tests were performed under the conditions described in Table 5.1. January 17, 1968, was clear and cold with moderate southwesterly winds in advance of a cyclone to the west. The ground was covered with snow up to 15 cm deep from a previous storm. During August 20–21, 1968, the dominant weather influence was a large anticyclone over the southeastern states. The day was hot, and the night was warm and humid. Skies were clear except for scattered afternoon cumulus.

Tests 1 and 3 were conducted between the time of afternoon maximum temperature and sunset. This is a period of neutral stability marking the transition from afternoon unstable to evening stable atmospheric conditions. At this time, considerable mixing of effluent from the 91-meter stack to the ground is expected after it travels over the bluff. The sampling sites for tests 1 and 3 were at the plume centerline, according to the wind direction trace at the 92-m level in the tower.

Test 2 was conducted after midnight, during stable conditions that were indicated by the rate of temperature and wind speed increase with height. Effluent from the Dresden stack would be expected to spread moderately in the horizontal plane but very slowly in the vertical plane, remaining mostly at the effective emission height. Wind direction data at the 92-m level showed 5- to 10-degree oscillations of variable periodicity. The mean wind direction, i.e., plume centerline, was about 4° (100 m) from the sampling point.

5.2.4 *Estimation of radionuclide concentrations and exposure rates*. Concentrations of ^{133}Xe and ^{135}Xe at the measurement locations were estimated from the

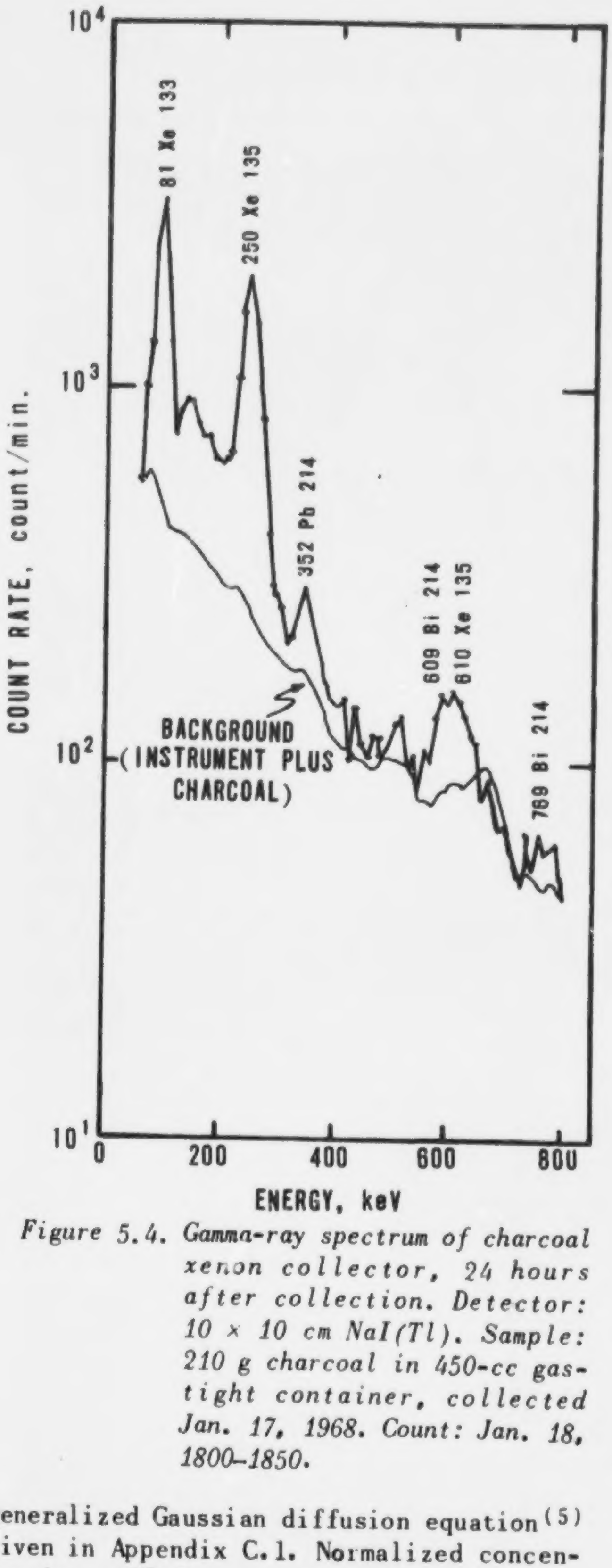

Figure 5.4. Gamma-ray spectrum of charcoal xenon collector, 24 hours after collection. Detector: 10 × 10 cm NaI(Tl). Sample: 210 g charcoal in 450-cc gas-tight container, collected Jan. 17, 1968. Count: Jan. 18, 1800–1850.

generalized Gaussian diffusion equation(5) given in Appendix C.1. Normalized concentrations (Xū/Q, see Appendix C.1) as a function of distance from the stack from extensive studies at Brookhaven(6) were used, except for vertical dispersion under stable conditions, for which Hanford studies provide the most realistic estimates.(7) The effective stack height was calculated by the Holland formula(8) as modified by Moses(9); however, the effective height was reduced by one-half of the elevation of the bluff above the base of the stack, i.e., 15 m. Release rates for ^{133}Xe and ^{135}Xe on January 17 and August 21 are given in Table 4.1.

To estimate the concentration of ^{138}Cs at the measurement site, the release rate at the stack of its parent ^{138}Xe was assumed to be 2.1 times the ^{135}Xe release rate (see Appendix B.3). The fractional ingrowth of ^{138}Cs five minutes after release was computed to be 9.4 percent of the ^{138}Xe release value, and the same normalized concentrations were used as for ^{133}Xe and ^{135}Xe.

The model for estimating radiation exposure rates is similar in approach to that described in the FSAR for Dresden 2 and 3(10) and that by Stuart and May,(11) with the following modifications: (1) dose calculations were made for photon energies of 0.4 and 2.0 MeV to represent by 2 average values all photons of energies less than and greater than 1 MeV, respectively; (2) dispersion of the stack plume was calculated as described above, based on a 1-hour sampling for all meteorological and plume-related measurements; and (3) no cross-wind averaging was done; exposures were computed for specific plumes. The estimated exposure rates are based on a 45-minute period for test 1, 10-minute averaging times for test 2, and a 30-minute period for test 3.

Calculation of these exposure estimates is shown in Appendix C.2. The gamma-ray energies and intensities that were averaged in terms of 0.4- and 2.0-MeV gamma rays are listed in Appendices C.3 and C.4. The Holland method(12) was used to check the estimation technique, as shown in Appendix C.5.

5.2.5 *Results and discussion.* Under the neutral conditions of tests 1 and 3, ^{133}Xe and ^{135}Xe were readily detected in the charcoal trap, as shown in Table 5.2 and Figure 5.4. For the described collection and detection conditions, these radionuclides

Table 5.1

Test Conditions for Sampling DNPS Stack Effluent in Environment

Test No.	1	2	3
Date	Jan. 17, 1968	Aug. 21, 1968	Aug. 21, 1968
Time	1620–1705 CST	0208–0258 CDT	1816–1846 CDT
Atmospheric stability classification	neutral	stable	neutral
Distance from stack, km	1.8	1.6	1.8
Sampling point azimuth, degrees	012	045	039
Wind direction, degrees			
92-m level	195	229*	220
38-m level	190	210–230	230–212
Wind speed, m/sec			
92-m level	8.5	6.7	6.2
38-m level	5.8	3.1	4.5
Temperature difference between 120 m and 10 m, °C	−0.5 to −0.2	+1.3	−1.1 to −0.7
Plume travel time, stack to sampler, min	3.5	4	5

*Average of 10-min values which ranged from 225 to 233 degrees.

are detectable at nominal gaseous fission product release rates as low as 1000 μCi/sec. Gamma-rays from short-lived ^{138}Xe and its ^{138}Cs daughter appear in the spectrum taken immediately after collection (Figure 5.2), but not clearly enough for analysis.

Under the neutral conditions in test 3, the air filters collected short-lived ^{138}Cs that could be measured with high precision 15 minutes after collection (see Figure 5.3). Interference by naturally occurring progeny of ^{222}Rn limits the sensitivity of this analysis, but was not serious at the indicated levels of ^{138}Cs. No radionuclides were found on the filter after 24 hours. Collection of the particulate daughter of ^{138}Xe thus is an alternative to collecting ^{133}Xe and ^{135}Xe, but filters must be analyzed for ^{138}Cs within several minutes, compared to hours for ^{135}Xe and days for ^{133}Xe.

The average exposure rates in tests 1 and 3 were 40 and 24 μR/hr, respectively (see Table 5.2); the variability of the individual measurements is indicated by the respective ranges from 22 to 46 and from 12 to 36 μR/hr shown in Figure 5.5. The

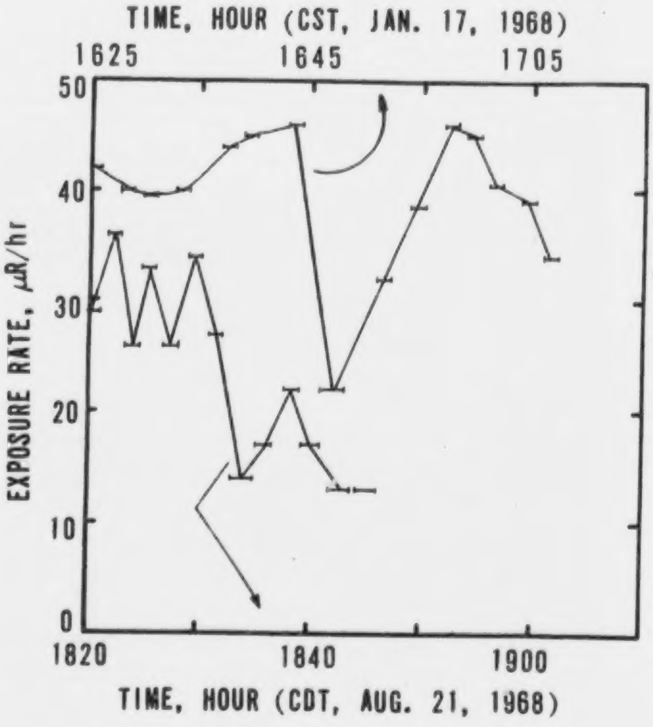

Figure 5.5. Exposure rates under neutral conditions, measured with muscle-equivalent ionization chamber.

Table 5.2
Radiation Exposure Rate and Radionuclide Concentration Measured in Air Near Ground Level Beneath Plumes from DNPS Stack

Test No.	1	2	3
Atmospheric stability	neutral	stable	neutral
Stack release rate, μCi/sec			
gaseous fission product release rate	15,000	11,600	11,600
^{133}Xe	400	730	730
^{135}Xe	1,660	1,420	1,420
Radiation exposure, μR/hr			
measured	40	13	24
calculated			
This study (App. C.2)	20	9	19
Holland (App. C.5)	28	9	29
Concentrations, nCi/m^3			
measured			
^{133}Xe	3.8 ± 0.1**	< 0.10	3.8 ± 0.1**
^{135}Xe	8.5 ± 0.4	< 0.17	7.0 ± 0.1
^{138}Cs	NS*	< 0.04	1.76± 0.02
calculated			
^{133}Xe (App. C.1)	1.0	0.003	2.2
^{135}Xe (App. C.1)	4.1	0.005	4.3
^{138}Cs	NS	0	0.9

*Not sampled

**Values are standard deviations in counting; < values are 3 σ of counting error.

Notes:

1. See Table 5.1 for dates, locations, and meteorological conditions.
2. Tests no. 1 and 3 were at centerline of plume; test no. 2 was 4° away from the centerline.

decrease toward the end of the measuring periods may be due to a shift in wind direction near the ground, a vertical movement of the plume center-line at the beginning of the transition from neutral to stable conditions, or a random fluctuation.

No radionuclides from the plume were detected at ground level under stable conditions in test 2 (Table 5.2). The vertical stability of the atmosphere, indicated by the rate of temperature and wind speed increase with elevation, presumably prevented dispersion of stack effluents to the ground. The gamma-ray spectrum of the charcoal (Figure 5.2) shows only a trace of background radiation from ^{222}Rn in air, and ^{226}Ra and ^{40}K impurities in the charcoal; that of the filters (Figure 5.3) shows only the short-lived airborne particulate progeny of ^{222}Rn.

The average exposure rate from radiation in the plume under stable conditions was 13 μR/hr, and individual values varied from 3 to 20 μR/hr (Figure 5.6). The measurement location, however, was not on the plume centerline. The exposure rate immediately

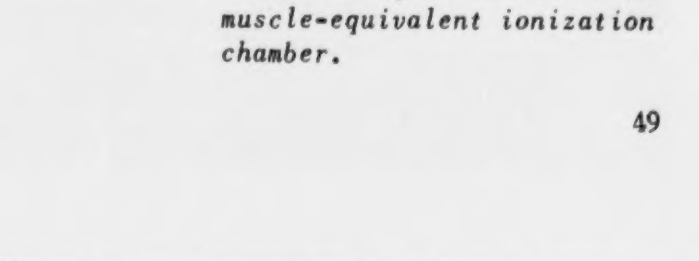

Figure 5.6. Exposure rates under stable conditions, measured with muscle-equivalent ionization chamber.

beneath the plume, approximately 4° distant from the measurement location, would be expected to be twice as high—i.e., 26 μR/hr. Exposure rate at the centerline of the plume therefore would be approximately the same under stable and neutral conditions.

The average measured ratio of the ^{133}Xe or ^{135}Xe concentrations at ground level 1.8 km from the stack under neutral conditions relative to their release rates at the stack (in terms of X/Q, as defined in Appendix C.1) was 6.2×10^{-6} sec/m^3; normalized for wind speed ($X\bar{u}/Q$), the average ratio was 4.7×10^{-5} m^{-2}. According to these ratios, atmospheric diffusion was lower—i.e., concentrations in environmental air were higher—than estimated from the Brookhaven data by approximately a factor of two. In view of the uncertainties associated with diffusion calculations, the two-fold difference should not be considered significant. Possible causes for the differences include: (1) slightly different wind directions at measurement locations than at the meteorological tower; (2) differences in wind speed with height; (3) a more complex effect of bluff height than was accounted for by the 15-m decrease in effective stack height in the estimates; (4) different ^{133}Xe or ^{135}Xe release rates during environmental measurements than during gas sampling at the station; (5) insufficient environmental concentration measurements for computing averages.

The average of the three measured external exposure rates was somewhat higher than the average of either of the two sets of calculated values; as indicated above, however, the differences are too small to be considered significant. Note that the results of the two sets of calculations themselves differ in two of the tests because of different approaches in computed exposure from a complex mixture of photon energies.

To indicate the utilization of such brief exposure measurements, an annual radiation exposure per unit release rate at the stack, $\bar{D}$, in μR/yr per mCi/sec, was estimated in 10° sectors from average short-term (1/2 to 1 hour) exposure rates, $\bar{D}'$, in μR/hr, and nominal gaseous fission product release rates, N, in mCi/sec, as follows:

$$\bar{D} = \frac{\bar{D}'}{N} \times 8760 \, \frac{\text{hr}}{\text{yr}} \times 0.75 \times \text{wind freq. to sector} \times \frac{\text{wind speed (during test)}}{\text{wind speed (avg. in sector)}} \qquad (5.1)$$

The factor 0.75 converts centerline exposure values to sector averages, according to the calculations by computer referred to in Section 5.2.4. The value of $\bar{D}'$ in test 2 must be multiplied by 2 to compensate for the off-centerline measurement. Values of $\bar{D}$, based on sector averages at a 46-m height of wind speed and frequency at Argonne National Laboratory,(1) are:

Test	Wind direction	Annual wind speed, avg.	Frequency	$\bar{D}$
1	190°	5.6 m/sec	0.041	1.1 $\frac{\text{mR/yr}}{\text{mCi/sec}}$
2	210°-230°	6.1	0.044	0.7
3	220°	6.2	0.040	0.5

From the average wind frequency and wind speed data,(1) the highest annual exposure is estimated for wind from the 210° sector (i.e., at a direction 30° east of north from Dresden); this estimated exposure is approximately one-fifth higher than for test 1, and one-third higher than for tests 2 and 3.

The average annual radiation exposure estimated from these three brief measurements in the three tests is 0.8 mR/yr per mCi/sec. At the 1968 annual release rate of 12,500 μCi/sec during 64 percent of the year, the exposure rate would be 6 mR/yr. This value can not be considered an accurate measure of the annual exposure rate at Dresden because the average short-term exposure rate is based on too few measurements and meteorological data were obtained at a distant location and at a height below that of the stack. The procedure suggests, however, that long-term external radiation exposures can be obtained from a sufficient number of short-term measurements of exposure rate, combined with the meteorological data that are now being collected at Dresden.

5.3 Long-term Radiation Exposure Rates

5.3.1 *Measurement.* TLD's were exposed for two periods at the 11 locations near Dresden shown in Figure 5.1, and at one background location 32 km NE of Dresden. The first exposure was between May 6 and 24, for 17 or 18 days, during refueling. The reactor was not in operation at this time and release rates of radioactive gases at the stack were undetectably low(13). The second period was for 14 days between August 15 and 29, during which the reactor was in operation for 12.2 days. The exposure rate at each station was measured with NaI(Tl) survey meters when the TLD's were placed in position and collected. Six dosimeters were exposed at station 110 to evaluate data reproducibility; duplicate dosimeters were placed at all other stations. Several stations were located in the prevailing direction of the plume toward northeast, and none was placed toward southwest, a relatively improbable plume direction.

5.3.2 *Analysis.* The TLD's (E,G&G model TL-12) were a powdered calcium fluoride-manganese mixture bonded to an electrical coil in an evacuated glass tube.* The dosimeters were annealed at 350°C at the Cincinnati laboratory on the day before exposure, and taken to the stations by car. After collection, the dosimeters were returned by car to the laboratory and sent by air mail to Las Vegas, Nevada, for reading. Five TLD's were annealed at the laboratory on the day before collection and sent to Las Vegas with the other dosimeters to measure the background during transportation to Las Vegas and storage at Las Vegas prior to reading.

The net radiation exposure of each TLD was computed from the gross reading by subtracting the above mentioned background value for shipment and storage, and also a constant internal background of 29 μR/hr. Gross readings were approximately 21 milliroentgen (mR), of which 17 mR was internal background, 1 mR, background from transportation and storage, and 3 mR, the net reading. The exposure rate, in μR/hr, was calculated by dividing the net exposure by the number of hours that each TLD had been exposed at the stations. To obtain the net exposure rate attributable to radiation from the Dresden plume, the average exposure rate at each station during refueling was subtracted from the average exposure rate at the same station during operation.

5.3.3 *Estimation of exposure rates.* Exposure rates from radiation in the plume at the eleven TLD stations between August 15 and 29 were estimated for comparison with measured values by the same computer techniques as for short-term exposures. The source term was taken to be an average gaseous fission product release rate of 11.6 mCi/sec during the 12.2-day period of reactor operation. Exposures were summed for the time intervals at which the plume passed near the TLD stations. Meteorological data were provided by the computer summary of Dresden tower readings, described in Section 5.1.2. The calculations are described in Appendix C.6, and an example is given in Appendix C.7. Estimated exposure rates and the plume rose (i.e., the wind rose rotated through 180°) for August 15 to 29 are shown in Figure 5.7. On the basis of wind direction frequency and wind speed, the highest exposure rate for the period was calculated to be 3.2 μR/hr in a direction 20° east of station 110.

5.3.4 *Results and discussion.* The TLD exposure rates for the two periods of measurement range from 5.3 to 12.7 μR/hr at the various stations (see Table 5.3), with an overall average of 9 μR/hr. This can be taken as an average background radiation exposure rate. Reproducibility of these dosimeter measurements, according to the standard deviation of six TLD's at station 110 and the mean standard deviations of duplicates at all stations, was approximately 1 μR/hr. The average exposure rate

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*We thank Mr. C. L. Fitzsimmons, Southwestern Radiological Health Laboratory, BRH, EHS, PHS, DHEW, for providing and reading these dosimeters.

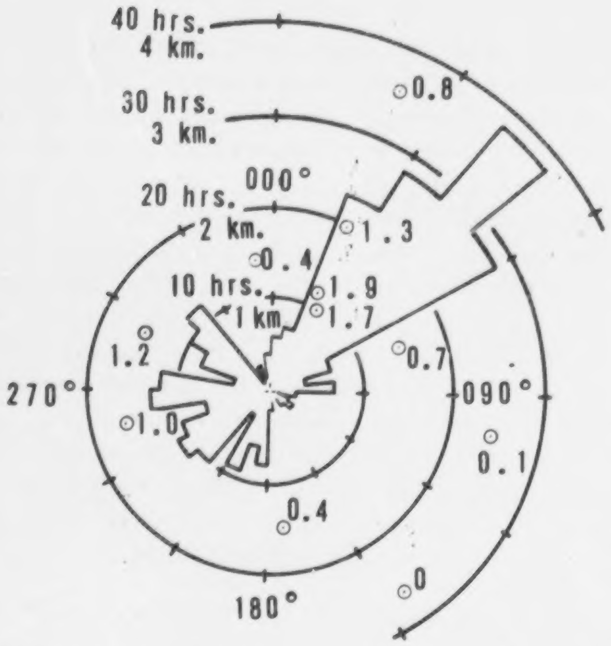

Figure 5.7. Plume rose and estimated exposure rates at dosimeter stations. Exposure rates in μR/hr. Numbers on circular grid indicate distance from Dresden in km. Occurrence of plume in indicated direction is shown in hours.

measured with pocket-type dosimeters by Dresden's contractor for environmental surveillance was 130 mR/yr (15 μR/hr).[14]

The differences in values between the two sets of exposure rates, attributed—at a first approximation—to radiation from the Dresden plume, are inconclusive. Improvements in techniques are necessary if long-term exposures are to be measured with TLD's at Dresden's current gaseous fission product release rates. Of the difference values in Table 5.3, three (#104, 106, and 110) are at exposure rates of 2 to 3 μR/hr and are larger than their standard deviation; all other exposure rates are less, and are smaller than their standard deviations. Note that three difference values are negative.

Although the values of individual measured exposure rates from radiation in the plume are uncertain, the sets of measured and estimated values are highly correlated, as shown in Figure 5.8. Lines of best fit have been drawn for all 11 values and for the 8 positive values; both lines

Table 5.3

External Radiation Exposure Rates Near Dresden
Measured with Powdered Calcium Fluoride TLD's*

Station[†] #	Measured Exposure Rate, μR/hr			Estimated Exposure Rate, μR/hr
	August 15-29	May 6-24	Difference	
101	8.3 ± 1.3[**]	7.9 ± 0.6[**]	0.4 ± 1.4	0.8
102	9.3 ± 1.3	8.0 ± 2.2	1.3 ± 2.6	0.1
103	9.4 ± 1.8	8.3 ± 1.4	1.1 ± 2.3	0.7
104	10.6 ± 1.1	8.6 ± 0.6	2.0 ± 1.2	1.3
105	9.9 ± 1.5	9.3 ± 1.4	0.6 ± 2.3	0.4
106	8.1 ± 1.3	5.3 ± 1.5	2.8 ± 2.0	1.2
107	7.7 ± 1.1	6.6 ± 0.0	1.1 ± 1.1	1.0
108	10.3 ± 2.8	12.7 ± 0.4	−2.4 ± 2.8	0.4
109	9.4 ± 1.1	10.5 ± 1.4	−1.1 ± 2.0	0.0
110	12.1 ± 0.9	8.9 ± 0.8	3.2 ± 1.2	1.9
111	9.2 ± 2.4	Lost	---	1.7
112	7.8 ± 1.5	9.9 ± 0.9	−2.1 ± 1.7	0.0

*EG&G Model TL-12

[†] See Fig. 5.1 for location

[**] ± values are 0.86 of range for duplicate dosimeters and standard deviation of sextuplicate readings at station #110.

Note: The reactor was in operation for 12.2 days between August 15 and 29, 1968; it was being refueled and was not in operation between May 6 and 24, 1968.

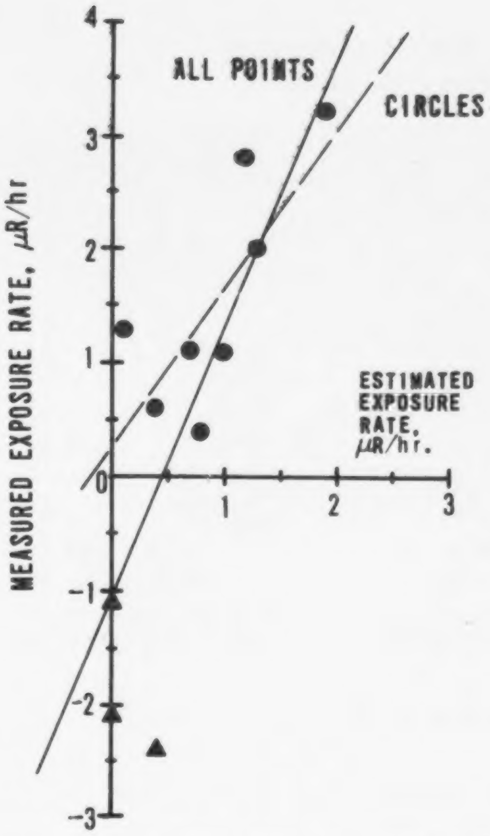

Figure 5.8. Comparison of measured and estimated exposure rates at TLD stations near Dresden on Aug. 15–29, 1968.

have a correlation coefficient of 0.77, and a slope that indicates that measured exposure rates near 2 μR/hr were approximately 1.5 times as high as estimated values.

In view of the significant correlation, further testing of the dosimeters for this purpose appears warranted. Annual external radiation exposures could thus be determined by summing biweekly or monthly measurements. To indicate the magnitude of the maximum annual exposure above background, the highest measured exposure rate, at station #110, was adjusted for annual wind-direction frequency and wind speed in the appropriate sector, and the annual average gaseous fission product release rate during 1968, as follows:

$$\bar{D}_{year} = \bar{D}_{meas} \times \frac{\text{frequency}_{year}}{\text{frequency}_{meas}} \times \frac{\text{speed}_{meas}}{\text{speed}_{year}} \times \frac{\text{release}_{year}}{\text{release}_{meas}}$$

$$= (3.2 \pm 1.2) \times \frac{0.042}{0.078} \times \frac{8.0 \text{ m/sec}}{5.9 \text{ m/sec}} \times \frac{0.64 \times 12{,}500\ \mu\text{Ci/sec}}{11{,}600\ \mu\text{Ci/sec}}$$

$$= 1.6 \pm 0.5\ \mu\text{R/hr}$$

$$= 14 \pm 5 \text{ mR/year} \qquad (5.2)$$

The extrapolation to annual exposure is based on meteorological data collected at Argonne National Laboratory, at a height of 46-m,[1] and is therefore not directly applicable to Dresden. It suggests, however, that the highest value will be measurable, but with considerable degree of uncertainty. Note that the uncertainty arises from the low exposure rate, and that exposure at higher rates would be accompanied by increased precision of measurement.

Whether differences in exposure rates between operating and non-operating periods may be attributed to radiation from the plume depends on the constancy of the natural radiation background. Although the negative difference values near stations 108 and 109 may be due to random error, they could alternatively result from a decrease in the background during the second exposure period. One indication of relative stability in background is the set of four exposure rate measurements with NaI(Tl) survey meters summarized in Table 5.4. With the single exception of station 101, differences among these measurements were less than the standard deviations for measurements with replicate TLD's. Only year-round tests will demonstrate whether one or even the average of several measurements of exposure rates from the natural radiation background can be subtracted from measurements during exposure from radiation in the plume to yield net exposures due to the plume at these low values.

In future studies, techniques can be improved by using TLD's with appreciably lower internal background, and by reading dosimeters near the stations to eliminate background exposure during transportation.

Table 5.4

External Radiation Exposure Rates from Natural Background Measured with Calibrated NaI(Tl) Monitors, μR/hr

Station #	Location, km	May 6 and 24	August 15 and 29
101	3.7 NNE	9.5 ± 0.8*	8.0 ± 0.2*
102	2.4 ESE	9.9 ± 0.1	9.9 ± 0.0
103	1.4 E	10.0 ± 0.2	10.1 ± 0.2
104	1.9 NNE	11.1 ± 0.2	10.6 ± 0.0
105	1.3 N	10.5 ± 0.0	10.1**
106	1.4 WNW	8.3 ± 0.1	7.9†
107	1.6 WSW	7.7 ± 0.4	7.7 ± 0.2
108	1.6 S	10.2 ± 0.6	10.1 ± 0.5
109	2.7 SE	9.4 ± 0.2	9.1 ± 0.1
110	1.3 NNE	10.7 ± 0.0	10.1 ± 0.1
111	1.1 NNE	6.9 ± 0.1	7.3 ± 0.4
112	32 NE	8.9 ± 0.4	8.9 ± 0.2

*values are 1/2 range of duplicate measurements obtained when dosimeters were exposed and when they were collected.

**value for August 29 only; centerline of plume from Dresden stack near station contributed to exposure rate on August 15.

†value for August 15 only; centerline of plume from Dresden stack near station contributed to exposure rate on August 29.

5.4 Portable Survey Meters

5.4.1 *Measurement*. Cylindrical 5 x 5 cm NaI(Tl) detectors with count-rate meters[*] were tested near Dresden for use as relatively sensitive and portable detectors of gamma radiation from the plume in the environment. To obtain readings, plumes were traversed by automobile, where roads permitted, in the directions indicated by instruments at the Dresden meteorological tower. The survey meters were also used to locate test sites at the centerline of the plume for detailed exposure-rate measurements, as described in Section 5.2.1. The NaI(Tl) survey meters are far more rugged and easier to read than the ionization chamber and Shonka electrometer, but provide only approximate exposure-rate values.

To attain even greater sensitivity, the plume was traversed with a cylindrical 10 x 10 cm NaI(Tl) detector on the roof of a light truck, 2.5 m above ground. The detector was covered with a 4-cm-thick urethane cover for thermal insulation. Gamma-ray spectra were analyzed with a 400-channel spectrometer within the truck. A gasoline-fueled alternator with Sola transformer provided power. Both types of instruments were frequently checked with radionuclide performance standards and by comparing background readings at fixed locations distant from the plume.

5.4.2 *Calibration*. The portable survey meters were calibrated for exposure-rate response by reading the Shonka electrometer at the same time. Calibration for radiation from the plume was obtained for a plume that had been released at the stack approximately 5 minutes earlier. The curve of count rate by survey meter *versus* exposure rate by ionization chamber ranged from 4 to 47 μR/hr above background as the plume shifted above the instruments. Calibration could be expressed in terms of the linear equation, Exposure Rate = m (Count Rate) + b, where the constants m and b depended on the voltage discriminator settings and varied among survey meters. Count rates of 20,000 counts/min

[*] We thank Mr. Richard Stoms, BRH, Cincinnati, for making these survey instruments and their background calibration, as well as the Shonka electrometers, available to us.

above background corresponded to exposure rates from the plume of approximately 20 μR/hr. Similar calibrations for the exposure rate from the natural radiation background had been obtained earlier by comparisons with the Shonka electrometer at a variety of locations. At the typical background exposure rate of 9 μR/hr, the count rate of the portable survey meters was 6,000 to 7,000 count/min.

Calibration of the large NaI(Tl) detector in terms of the concentration in air of gamma-ray-emitting radionuclides was attempted. The spectra obtained from plumes could not be analyzed, however, because comparison spectra were not available for short-lived radionuclides that contributed significantly to the total spectra (see Section 4.2.1).

5.4.3 *Results and discussion.* The plume from the Dresden stack was readily detectable by NaI(Tl) survey meters within a 5-km radius of Dresden. Examples of the response of the instrument for 2-minute measurements in Figure 5.9 demonstrate the sensitivity as well as the variation in exposure rate in traversing the plume. On the basis of readings at the centerline of the plume, the average environmental exposure rate of 9 μR/hr is approximately doubled at distances of 1.6 km by a stack release of 2 mCi/sec. The values in Figure 5.9 do not show a true profile of exposure rates in the plume, because the plume moves during the traverse of the plume with the survey meter. Exposure values based on the calibration are approximate, since the relative intensity of gamma-ray energies changes with the time of travel of the plume. The calibrations appear to be sufficiently accurate, however, to permit use of the instruments to indicate significant changes in radionuclide releases at the stack and potential health hazards.

The spectra obtained with the larger detector mounted on the truck roof, shown by the 10-minute measurements in Figure 5.10, indicate that the plume can be identified 15 km distant from the stack at release rates of 15,000 μCi/sec. This sensitivity should also be attainable without spectrometer, by counting in energy regions that have high count rates in the plume, compared to the natural radiation background. The count rate between 40 and 200 keV, for example, is 3,600 counts/min above background at the most distant point (spectrum 5 in Figure 5.10), compared to 16,000 counts/min background; a more favorable energy region according to Figure 5.10 would be 200–300 keV. The 10-minute average exposure rate measured with the portable survey meter at that location, 18 km NE of Dresden, was 2.5 μR/hr above background.

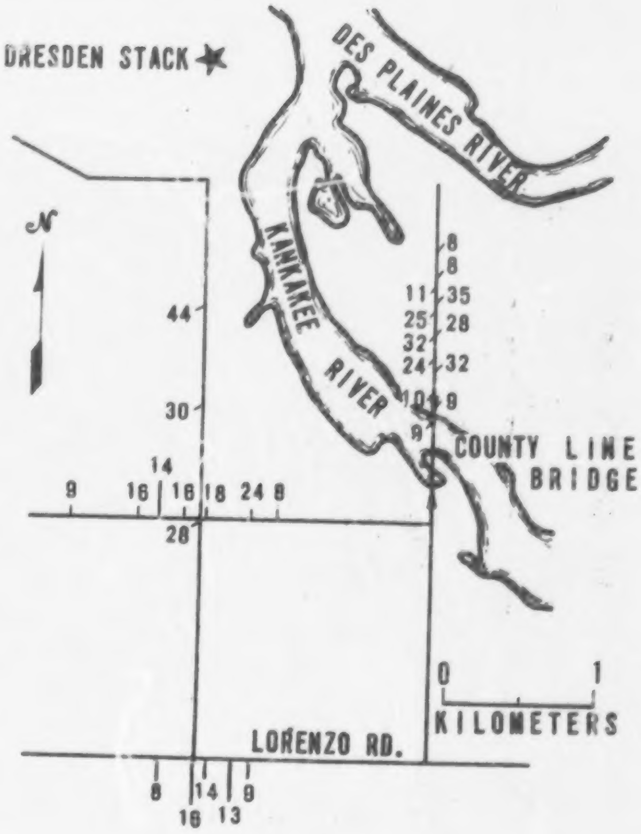

Figure 5.9. Typical exposure rates measured with portable NaI(Tl) survey meters. Values are in μR/hr above background; values south-east of stack were measured on Nov. 14, 1967, values to south on Nov. 15, 1967. All values were measured in plume from Dresden stack.

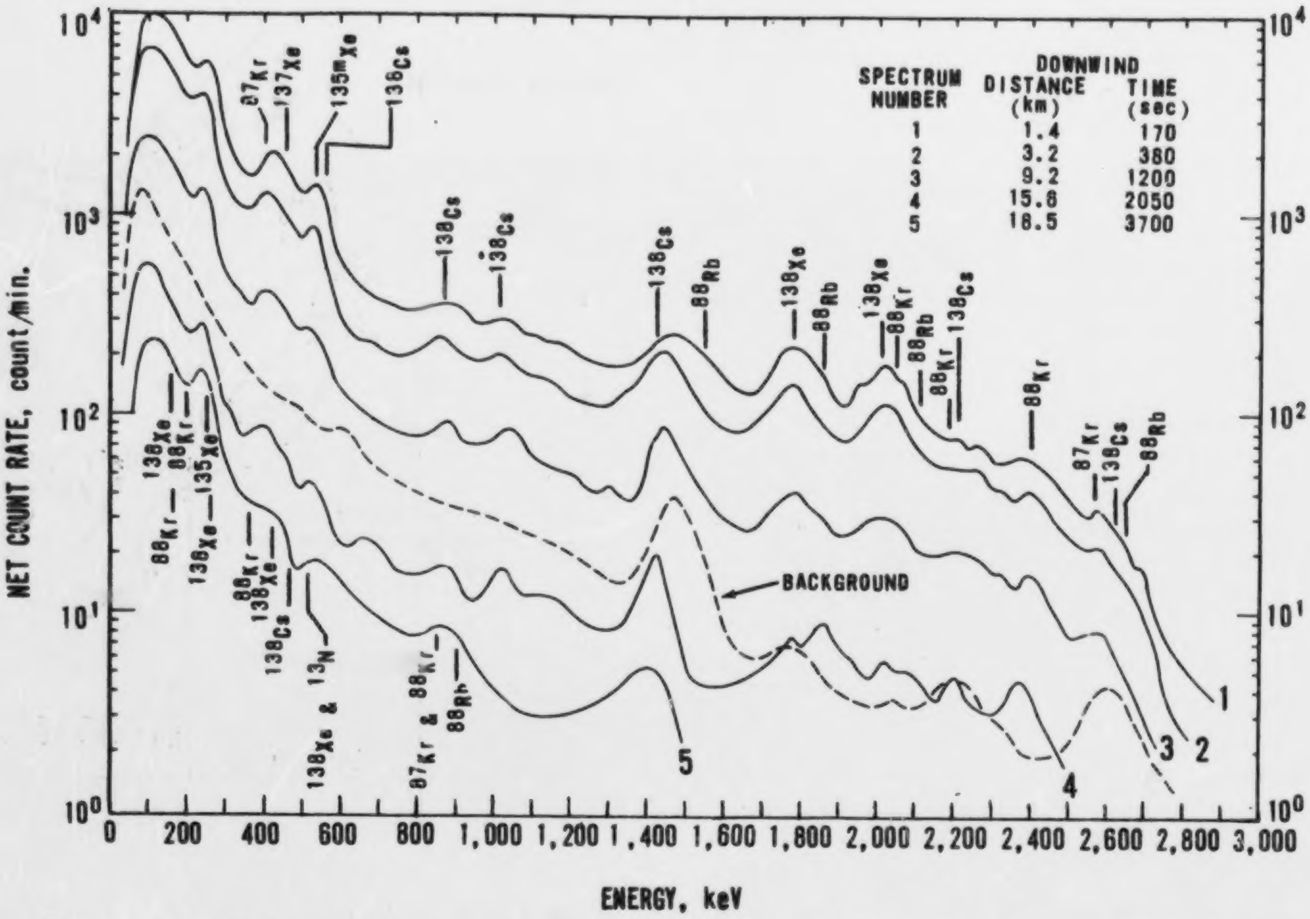

Figure 5.10. Net gamma-ray spectra measured on ground near center-line of plume with 10 × 10 cm NaI(Tl) detector.

5.5 References

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6. RADIONUCLIDES IN SURFACE WATER

6.1 Water Use in the Illinois River

6.1.1 *Public water supply and fishing.* Liquid wastes at Dresden are discharged into the coolant-water discharge canal. The canal empties into the Illinois River at the confluence of the Des Plaines and Kankakee Rivers at Illinois River Mile* (IRM) 272.4, just upstream from Dresden Dam at IRM 271.6 (see Figure 6.1). Flow rates at the Marseilles gaging station (IRM 246.6), which are only a few percent higher than at Dresden Dam,(1) for the period 1920 to 1963 were:(2)

Maximum daily – 94,000 cubic feet per second* (cfs) on July 14, 1957

Minimum daily – 1,500 cfs on October 16, 1943

Average – 10,900 cfs

Because its water has been polluted with domestic sewage and industrial waste from Chicago since the opening of the Sanitary

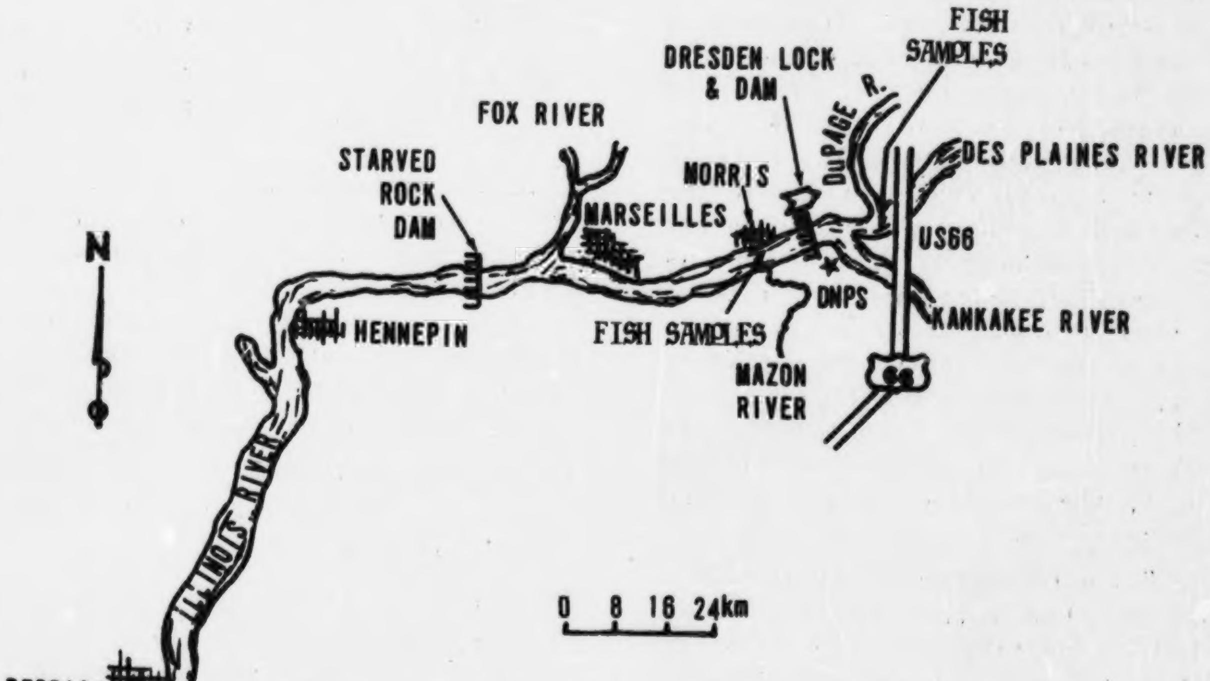
FISH SAMPLES
DRESDEN LOCK & DAM
DuPAGE R.
FOX RIVER
STARVED ROCK DAM
MORRIS
DES PLAINES RIVER
MARSEILLES
N
US66
HENNEPIN
FISH SAMPLES
DNPS
KANKAKEE RIVER
66
MAZON RIVER
ILLINOIS RIVER
0 8 16 24km
PEORIA

Figure 6.1. Illinois River below Dresden.

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*1 mile = 1.61 km; 1 cfs = 28.3 liters/sec.

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and Ship Canal into the Des Plaines River in 1900, the Illinois River is not used for public water supply, fishing, or irrigation near Dresden.

The fish in the river near Dresden consist primarily of carp, goldfish, shiner, and catfish.(3) Catfish that were ''fungused'' and otherwise in unwholesome condition were found in 1912, and malformed (knothead) carp, in 1928; both conditions were attributed to pollution.(3) According to inhabitants of Morris (IRM 262), the fish have a reputation for bad taste and odor, and only someone unfamiliar with the river would fish at this location. The only commercial fishing in the Illinois River above Hennepin (IRM 209) is by a single fisherman in the mouth of the Fox River (IRM 241).(3) Sport fishing begins below Starved Rock Dam (IRM 232).(1)

The first use of the Illinois River below Dresden as public water supply is at Peoria(1) (IRM 166). The Peoria water supply has three sources: ground-water wells, an infiltration gallery with induced recharge from the Illinois River, and direct intake from the Illinois River via a well for settling prior to treatment. Treatment is by pH control and chlorination. Ground-water wells are the major source of water; the infiltration pit is used only when the river is extremely turbid. The average dilution factor for radionuclides in the Illinois River at Peoria relative to the coolant-water discharge canal at Dresden is approximately 50, based on a flow rate at Peoria that was 1.6 times as great at Peoria as at Marseilles in 1961–1962(1) (i.e., 1.6 x 10,900 cfs = 17,000 cfs), and a flow rate in the canal of 370 cfs (166,000 gal/min). At maximum and minimum flow rates, the dilution factors are 400 and 6, respectively.

6.1.2 *Radiation exposure calculations.* Although Section 6.1.1 indicates that neither drinking water nor food fish are obtained at the mouth of the coolant-water discharge canal, it is of interest to estimate the potential radiation exposure from radionuclide discharged by Dresden at that point. For drinking water, the average radionuclide concentrations measured on three occasions for ionic substances and on two occasions for insoluble substances compare as follows to AEC limits:

Radionuclide		Net avg. conc. in coolant-canal discharge water,* pCi/l	Limit** pCi/l	Concentration / Limit
^{3}H	(S)	8.	3×10^{6}	3×10^{-6}
^{58}Co	(S)	1.0	1×10^{5}	1×10^{-5}
	(I)	0.2	9×10^{4}	2×10^{-6}
^{60}Co	(S)	0.4	5×10^{4}	8×10^{-6}
	(I)	0.4	3×10^{4}	1×10^{-5}
^{89}Sr	(S)	2.5	3×10^{3}	8×10^{-4}
	(I)	0.01	3×10^{4}	3×10^{-7}
^{90}Sr	(S)	0.2	3×10^{2}	7×10^{-4}
	(I)	0.03	4×10^{4}	8×10^{-7}
^{131}I	(S)	6.	3×10^{2}	2×10^{-2}
	(I)	< 0.02	6×10^{4}	$< 4 \times 10^{-7}$
^{134}Cs	(S)	0.5	9×10^{3}	6×10^{-5}
	(I)	0.05	4×10^{4}	1×10^{-6}
^{137}Cs	(S)	1.4	2×10^{4}	7×10^{-5}
	(I)	0.3	4×10^{4}	8×10^{-6}
^{140}Ba	(S)	1.4	3×10^{4}	5×10^{-5}
	(I)	0.05	2×10^{4}	2×10^{-6}
sum				2×10^{-2}

*Differences between concentrations in discharge and intake water; averages of 3 values in Table 3.3 for soluble (S) fraction; averages of 2 values in Table 3.4 for insoluble (I) fraction; ^{3}H concentration based on analysis of liquid waste before discharge (see Section 3.3.7).

**Limiting annual average in 10 CFR 20, Appendix B, Table II, Column 2; 1 pCi/l = 1 x 10^{-9} μCi/ml.

All radionuclides except ^{131}I are below the indicated concentration limits for drinking water by factors exceeding 1,000; the relatively high ^{131}I ratio derives from the single high value measured on January 17–18, 1968. Under typical conditions of dilution in the Illinois River (50-fold) and mixing with well-water at Peoria (4-

fold), radionuclide concentrations in drinking water at Peoria would be approximately 200-fold lower than at the point of discharge by Dresden. A further decrease in the concentration of some radionuclides in water would be expected through mechanisms such as radioactive decay, uptake by aquatic organisms, or retention on benthal deposits in the 106-mile passage to Peoria.

To evaluate potential radiation exposure from eating fish that had lived at the point of discharge of liquid effluent at Dresden, the amount of fish was estimated that would correspond to the consumption of 2,200 ml of water at AEC limits:

Radionuclide	Concentration factor,* l/kg	Hypothetical concentration in fish, μCi/kg wet wt.	Intake limit for fish corresponding to limit for water,** kg(wet wt.)/day
^{3}H	0.9	7 x 10^{-6}	1 x 10^{6}
^{58}Co	500	6 x 10^{-4}	400
^{60}Co	500	4 x 10^{-4}	300
^{89}Sr	40	1 x 10^{-4}	70
^{90}Sr	40	9 x 10^{-6}	70
^{131}I	1	6 x 10^{-6}	100
^{134}Cs	1,000	6 x 10^{-4}	30
^{137}Cs	1,000	2 x 10^{-3}	20
^{140}Ba	10	1 x 10^{-5}	7,000

*Assuming density of 1 kg/l in fish flesh.

**At water intake of 2.2 liter/day; i.e., weight of fish in kg/day = 2,200 ml/day x limit in μCi/ml ÷ concentration in fish in μCi/kg; limit from 10 CFR 20, Appendix B, Table II, Column 2, for soluble substances.

To compute the hypothetical radionuclide concentrations in fish flesh, the soluble and insoluble concentrations measured in coolant-canal water were added for each isotope, and the sum was multiplied by the concentration factor for each element in fresh-water fish.(4)

The critical radionuclides by this pathway are ^{137}Cs and ^{134}Cs, at daily individual consumption of 20 and 30 kg of fish, respectively. For all of the listed radionuclides, the limit for continuous daily consumption is 9 kg of fish, which is considerably more than the reported daily average intake by fishermen of 0.05(5) or 0.1(6) kg. Radionuclide concentrations in fish that are caught farther downstream would be expected to be lower because radionuclide concentrations in downstream water are undoubtedly lower. On the other hand, the above estimate does not consider the possible concentration of radionuclides in the food chain between water and the food fish, i.e., in aqueous biota or fish that are consumed by the food fish. Moreover, intake of radionuclides with other foods reduces the amount of radioactivity that may be consumed with fish.

6.2 Water at Peoria, Illinois

6.2.1 *Sampling and analysis.* Radionuclides in ionic form were collected with the effluent sampler described in Section 3.1.4 from two sources on May 6-7, 1968:

Sample	Volume, liters	Collection period, hours	Hardness as $CaCO_3$, mg/liter
settled, untreated river water	52	18	320
drinking water	100	18	340

In addition, 1-liter aliquots of water were collected to measure hardness and tritium. The effluent samplers were operated in the Peoria Water Treatment Plant with the aid of plant staff.* During the sampling period, 22 percent of the water supply was from the river and 78 percent from ground-water wells. The infiltration pit was not used.

Procedures described in Section 3.2.2 were followed, except that the ion-exchange resins were not washed with distilled water to remove silt. Tritium was measured with a less sensitive liquid scintillation system and was not found at a minimum detectable level of 2 pCi/ml. Radiocobalt and radiocesium concentrations were too low to distinguish between ^{58}Co and ^{60}Co, and between ^{134}Cs and ^{137}Cs, respectively.

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*We thank Messrs. J. Behee and A. Cherry, Peoria Water Treatment Plant, for helping to collect these samples and describing the plant operation.

6.2.2 *Results and discussion.* River water at Peoria contained radiocobalt and ^{90}Sr at concentrations of approximately 1×10^{-9} μCi/ml each, and no detectable ^{89}Sr or ^{131}I, as shown in Table 6.1. Strontium-90 concentrations of 1.5, 1.1, and 1.1 pCi/liter, measured at Peoria by the Federal Water Pollution Control Administration(7) in September 1967, March 1968, and September 1968, respectively, agree with the listed ^{90}Sr value. The Dresden environmental monitoring contractor reported a ^{90}Sr concentration of $0.8 \pm 0.3 \times 10^{-9}$ μCi/ml, and an ^{89}Sr concentration of $2.7 \pm 0.8 \times 10^{-9}$ μCi/ml, for the May 1-15 composite sample of Illinois River (Dresden Dam) water.(8) All radionuclide concentrations were well below the AEC limits listed in the last column of Table 6.1.

The radiocobalt and possibly part of the radiocesium and ^{90}Sr are attributed to sources other than fallout. Concentrations of ^{90}Sr and ^{137}Cs from fallout in Kankakee River water (Dresden coolant-water intake canal) averaged respectively 0.4×10^{-9} and 0.2×10^{-9} μCi/ml in January, June and August, 1968 (see Table 3.3). Dresden, Argonne National Laboratory,(9) and sewage and industrial waste from Chicago may have contributed the radioactivity that did not originate in fallout.

As shown in Table 6.1, radionuclide concentrations in drinking water were lower than in river water for all measured radionuclides. This can be accounted for by the almost five-fold dilution of river water with ground water. The latter would be expected to contain no radiocobalt, and less ^{90}Sr and ^{137}Cs than Illinois River water.

6.3 Fish at Dresden

6.3.1 *Sampling and analysis.* Fish were caught with electro-shocking equipment* at the 2 locations shown in Figure 6.1 on June 27, 1968. The following fish were collected:

Location	Type	Number	Wet weight
Illinois River (IRM 262)	carp	11	11.8 kg
	shad	10	1.0
Des Plaines River (1 mi. W of highway US 66 bridge)	carp	7	5.0
	goldfish	30	5.4

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*We thank Messrs. W. Starrett, D. Dooley, and J. Stalter, Natural History Survey Division, State of Illinois, for collecting these samples.

Table 6.1

Radionuclide Concentration at Peoria, Ill., Water Treatment Plant May 6-7, 1968, pCi/liter*

Radionuclide	River water	Drinking water	Permissible concentration†
^{58}Co + ^{60}Co	1.0 ± 0.3	0.3 ± 0.1	5×10^{4} (^{60}Co)
^{89}Sr	< 0.1	< 0.1	3×10^{3}
^{90}Sr	1.1 ± 0.2	0.7 ± 0.1	3×10^{2}
^{131}I	< 0.1	< 0.1	3×10^{2}
^{134}Cs + ^{137}Cs	1.5 ± 0.2	0.3 ± 0.5	9×10^{3} (^{134}Cs)

*Concentration at sampling; pCi/liter = 1×10^{-9} μCi/ml.

†For soluble substances in effluents to unrestricted area, 10CFR20, Appendix B, Table II, Column 2 (1965).

Note: ± values are 1 σ counting error; < values are 3 σ counting errors.

Fish from the Des Plaines River, 4.5 miles upstream from Dresden, were obtained to determine background values attributable to fallout from nuclear weapon tests, sewage and waste from Chicago, or discharge by the Argonne National Laboratory. The downstream sample was collected below Dresden Dam, approximately 10 miles from the coolant canal discharge point.

Fish were collected just below Dresden despite the absence of fishing in this region on the assumption that these fish would accumulate at higher concentrations the same radionuclides retained by fish farther downstream. To minimize confusion due to the movement of individual fish upstream, downstream, or into tributaries, all samples were composited for analysis.

Samples were frozen immediately after collection. For analysis, the fish were weighed, thawed, and dissected to separate tissues that were expected to concentrate the radionuclides of interest:

heart area
(for thyroid) – ^{131}I analysis
muscle – ^{134}Cs and ^{137}Cs analysis
kidney – ^{58}Co and ^{60}Co analysis
liver – ^{58}Co and ^{60}Co analysis
bone – ^{89}Sr and ^{90}Sr analysis

The tissues were combined by type of fish. Heart area, liver, and kidney were analyzed directly by gamma-ray spectrometry with a 10 x 10 cm NaI(Tl) detector. Muscle was ashed at 400°C and then analyzed by gamma-ray spectrometry. Muscle, kidney and liver samples were also analyzed with the gamma-ray coincidence/anticoincidence system (see Section 3.2.2). Bone samples were ashed at 600°C, strontium was separated chemically, and radiostrontium and ^{90}Y were measured with low-background beta counters. Stable strontium and calcium in bone were determined chemically, and potassium in muscle was determined by gamma-ray spectrometric measurement of ^{40}K.

6.3.2 *Results and discussion.* Concentrations of ^{90}Sr and ^{137}Cs in fish are summarized together with stable potassium, strontium and calcium levels in Table 6.2. Only the radionuclides ^{137}Cs and ^{90}Sr were in the fish in detectable amounts. No significant concentration differences were observed between fish collected above and below the nuclear power station. The average ^{137}Cs concentration in the muscle, weighted according to the number of fish in each group, was 14.5 pCi/kg wet weight, and the average ^{137}Cs/K ratio was 6.9 pCi/g K. The average ^{90}Sr concentration measured in the bone, weighted according to number of fish in each group, was 500 pCi/kg fresh weight, 4.1 pCi/mg Sr and 6.3 pCi/g Ca. The average strontium concentration was 0.12 g/kg.

Reported concentrations of ^{137}Cs and ^{90}Sr in fresh-water fish vary greatly; both range from about 20 to 20,000 pCi/kg fresh weight.(10,14,16,17) The variations are due to a number of factors, including:

1. the concentration of the radionuclide in the aquatic environment;

Table 6.2

Radionuclide (pCi/kg)* and Stable Ion (g/kg)* Concentration in Fish Tissue
June 27, 1968

Tissue	Substance	Des Plaines Carp	Des Plaines Goldfish	Illinois Carp	Illinois Shad
Muscle	^{137}Cs	14.3 ± 0.5†	11.1 ± 1.1	9.6 ± 0.4	31 ± 3
	Potassium	1.9	2.3	1.8	2.3
Bone**	^{90}Sr	590 ± 60	420 ± 50	590 ± 80	550 ± 30
	Strontium	0.14	0.14	0.11	0.08

*All kg values are wet weight.

**Calcium content = 78 g/kg.

†± values are 2 σ counting error.

2. exposure frequency;(13)

3. the presence of other elements; for example, the uptakes of stable strontium and ^{90}Sr are affected by the calcium content of the water;(10,12,17)

4. the effect of temperature, because metabolic processes of cold-blooded aquatic organisms generally become slower as the water temperature decreases;(13) and

5. the trophic level. Predatory fish nearly always contain greater concentrations of ^{137}Cs and ^{90}Sr than non-predatory fish. For example, Kolehmainen, *et al.*(11) observed an average predatory/non-predatory ratio of 3.8 for ^{137}Cs; Gustafson, *et al.* reported a ratio of 3.5 for pike *vs.* perch (the perch being largely consumed by pike).(16)

Although ^{90}Sr was not measured in the scales of the Dresden fish, it is interesting that Ruf has reported nearly identical concentrations in fish bone and scales, and similar ^{137}Cs concentration in meat and bone.(15) Similarly, Ophel and Judd found concentrations of Sr and Ca to be similar in bone, scales and gills.(18)

Another approach to evaluating concentrations is in terms of the accumulation factor (AF)—the ratio of wet-weight concentration in fish to concentration in water. Water analyses by the U.S. Public Health Service and the Federal Water Pollution Control Administration in the Illinois River at Peoria(7) confirm the hardness value in Section 6.2.1, which indicates a calcium concentration of 130 mg/liter. They also show average strontium concentrations (1962 to 1968) of 180 μg/liter. Relative to these stable-ion concentrations and the ^{90}Sr and radiocesium concentrations (assuming that all radiocesium is ^{137}Cs) in Table 6.1, the average concentrations in fish yield the following accumulation factors:

$AF_{^{90}Sr} = 450$ $\quad$ $AF_{Ca} = 600$

$AF_{^{137}Cs} = 10$ $\quad$ $AF_{Sr} = 690$

AF values are in meat for ^{137}Cs and in bone for strontium and calcium.

Accumulation factors for strontium in bone have been reported to be 100–4030 for perch,(17) 50–8810 for pike,(17) 70–9170 for roach,(17) 145–10,000 for brown trout,(19) and an average of 3940 for crappie.(14) The AF_{Sr} for skin and scales of fish appear to be about the same as for bone.(17) For ^{90}Sr, Ruf found values between 200–600 for bone of carp, trout and pike.(15) The average $AF_{^{90}Sr}$ and AF_{Sr} for the Dresden fish fall within their respective ranges, and there is no significant difference between these two values, or between them and the AF_{Ca}. The accumulation factor reported for ^{137}Cs varies greatly, being 232–15,000 for perch, 195–11,000 for pike, 122–4,850 for roach, 122–895 for bream and 990–1,920 for whitefish.(12,16) The $AF_{^{137}Cs}$ observed in the Dresden fish is small compared to these observed ranges. In the presence of Chicago sewage, however, the Illinois River does not provide a typical aqueous environment for fish at the sampling points.

A third basis for comparison is the observed ratio for ^{90}Sr—in case of fish, the $OR_{bone/water}$ for strontium relative to calcium, computed by dividing the $AF_{^{90}Sr}$ by the AF_{Ca}. In Dresden fish, the $OR_{bone/water}$ is 450/600 = 0.8 for ^{90}Sr and 690/600 = 1.2 for stable strontium. A range of values would be expected because of fluctuations in the concentrations of ^{90}Sr, strontium and calcium during the lifetime of the fish. Reported observed ratios are 0.2 to 0.7 for strontium(10,15,18) and 0.1 to 0.4 for ^{90}Sr.(15) The calculated $OR_{bone/water}$ for strontium and ^{90}Sr in Dresden fish are thus higher than the ranges listed in the literature.

No other radionuclides were found in fish. Minimum detectable concentrations set by sample size and sensitivity of radiation detector were:

Tissue	Radio-nuclide	Carp and goldfish	Shad
muscle	^{134}Cs	1 pCi/kg	3 pCi/kg
bone	^{89}Sr	1,000	1,000
kidney	^{58}Co	100	500
	^{60}Co	80	300
liver	^{58}Co	9	40
	^{60}Co	7	25
heart area	^{131}I	300	800

Shad was analyzed with poorer sensitivity because the sample was small. Increased sensitivity is desirable in future analyses for ^{89}Sr and ^{131}I.

6.4 References

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7. RADIONUCLIDES IN THE TERRESTRIAL ENVIRONMENT

7.1 Dresden Environment

7.1.1 *Site*. The Dresden Nuclear Power Station is located on a 3.9-km^2 (953-acre) tract in Goose Lake Township, Grundy County, Illinois, on the shore of the Illinois and Kankakee Rivers (see maps in Figures 7.1 and 7.2). The plant is on rolling prairie that is readily drained; bedrock is within four feet of the surface. Plant elevation is 155–160 m. The only higher area near the plant is Kankakee Bluffs at elevation 180–190 m to the north and east of Dresden on the far shore of the Illinois River.

An exclusion distance of 0.8-km radius surrounds Dresden. Dresden II and III plants are under construction immediately to the west of Dresden I and will have similar exclusion distances. The Midwest Fuel Reprocessing Plant of the General Electric Company is being built 2 km south of the Dresden Nuclear Power Station complex. Surrounding Dresden are small farms growing field corn, pasture grass for grazing beef cattle, and, to a lesser extent, soybeans and winter wheat. Eleven relatively small dairy herds—some of only 2 to 4 cows—were seen at distances between 3 and 20 km from Dresden. Wooded areas consist of the McKinley (Channahon) State Forest Preserve on Kankakee Bluffs, and the Des Plaines Wildlife Conservation Area, east of Dresden between highway US 66 and the Kankakee Cutoff. There are several abandoned strip mines 10 km south of Dresden, and a small mine operates 3 km south. In addition to the Kankakee, Des Plaines, and Illinois Rivers, there are small streams in the neighborhood, including the Illinois and Michigan Canal, the DuPage River emptying into the Des Plaines River 5 km NE, the Aux Sable Creek flowing south into the Illinois River 5 km W, and the Mazon River flowing north to the Illinois River 11 km SW of Dresden. The entire area is accessible by highways and dirt roads that generally lie on a 1-mile (1.6 km) grid pattern.

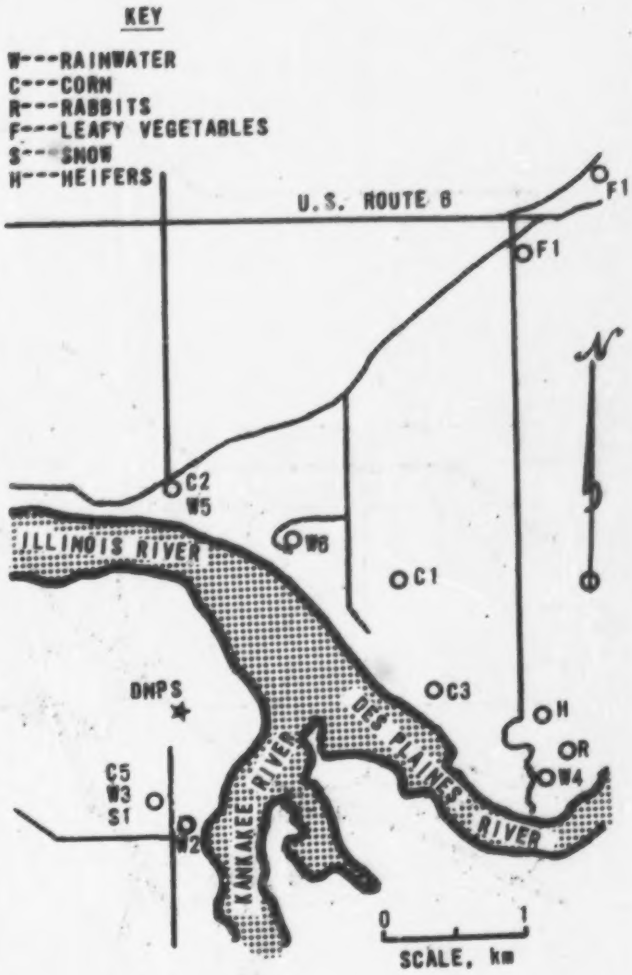

Figure 7.1 Sampling locations in immediate vicinity of Dresden Nuclear Power Station.

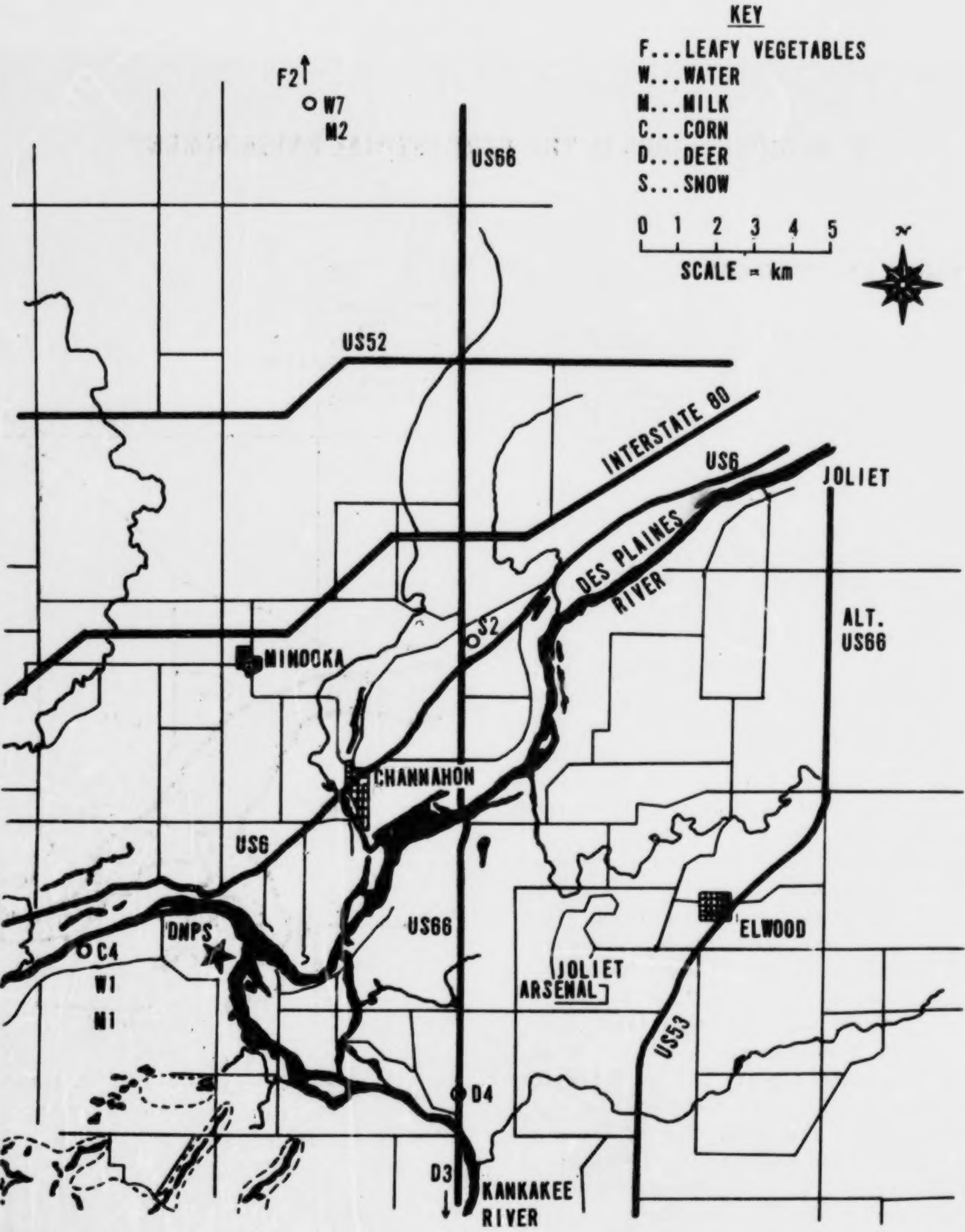
KEY
F...LEAFY VEGETABLES
W...WATER
M...MILK
C...CORN
D...DEER
S...SNOW
0 1 2 3 4 5
SCALE = km
N
F2
W7
M2
US66
US52
INTERSTATE 80
US6
JOLIET
DES PLAINES
RIVER
ALT.
US66
S2
MINOOKA
CHANNAHON
US6
US66
DNPS
C4
W1
M1
ELWOOD
JOLIET
ARSENAL
US53
D4
D3
KANKAKEE
RIVER

Figure 7.2. Sampling locations in general vicinity of Dresden Nuclear Power Station.

The nearest homes are cottages 1.2 km SSE of Dresden along the west bank of Kankakee River, 2 houses on the Kankakee Bluffs approximately 1.4 km NE of Dresden, and the operating station at the Dresden Dam and Locks, 1.4 km NW of Dresden. Within a 5-km radius, there are numerous cottages on the island SE of Dresden formed by the Des Plaines River, Kankakee River and Kankakee Cutoff, and scattered farm houses. The town of Channahon (pop. 1,200), 5 km NE of Dresden, is the largest population center near Dresden. Within 13 km of Dresden are Morris (pop. 7,900) to the west, and the smaller towns of Coal City and Wilmington, S and SE, respectively. Large cities in the vicinity of Dresden are Joliet (pop. 67,000), 20 km NE, and Aurora (pop. 64,000), 40 km N. Chicago is 80 km NE of Dresden.

The largest industrial complex near Dresden is the Joliet arsenal, 9 km E. The north shore of the Illinois River is being developed as an industrial area, and there are factories along highway US 66, 6 km to the east.

7.1.2 *Environmental surveillance.* It was the purpose of this study to demonstrate and test sensitive and informative monitoring techniques rather than to parallel the extensive surveillance program that is carried out at Dresden. Emphasis was placed on collecting foods (as direct sources of radiation exposure to humans) or indicators of radioactivity concentrations in foods, measuring even very low concentrations of radionuclides rather than reporting less-than values, and comparing measured concentrations with estimated values. Estimates were based on release rates at the Dresden stack, meteorological data, diffusion calculations, and environmental and biological transfer coefficients. The study criteria were met in several instances, but further efforts are required to measure very small amounts of radionuclides in a number of media.

Environmental surveillance that provides specific information about radionuclide concentrations is difficult at many nuclear facilities, including Dresden, because release rates of radionuclides to the air are low, and radionuclides in fallout from atmospheric nuclear tests interfere with analyses. Of the radionuclides discharged at the Dresden stack, ^{3}H, ^{90}Sr, and ^{137}Cs are also continuously deposited in fallout; the presence and relative concentrations of ^{131}I, ^{140}Ba, and ^{89}Sr in fallout depend on the time interval between the tests in which the fallout radionuclides were formed and sample analysis. Short-lived radionuclides in fallout during the study period were derived from the mainland Chinese atmospheric nuclear test on December 24, 1967. Other possible, although unlikely, sources were leakage from underground and cratering tests in Nevada on Jan. 18, Jan. 26, and March 13, 1968, and the French atmospheric nuclear tests in the South Pacific on July 7, July 15, and Aug. 3, 1968.

Radionuclides that could be attributed to Dresden stack effluent if found in the immediate neighborhood of Dresden include the short-lived noble gases, their short-lived progeny, and ^{58}Co and ^{60}Co; these are released at the Dresden stack (see Section 4.1) and are usually not detected in fallout from nuclear tests. The short-lived noble gases and their short-lived progeny do not remain long in environmental samples, however, and radiocobalt is released at the stack in extremely low amounts (see Table 4.6).

As an alternative approach, radionuclide concentrations were compared in samples collected near the stack and at a distance from it, or in the downwind direction *versus* upwind. Higher concentrations of indicator radionuclides in the nearby or downwind samples may be attributed to discharge at the Dresden stack. Strontium-89 and ^{131}I were selected as indicator radionuclides for deposition, because their release rates were highest among stack effluents other than noble gases and tritium. The release rate of ^{89}Sr in stack effluent is actually higher than measured at the stack because it is formed in the ^{89}Kr-^{89}Rb-^{89}Sr decay chain to the extent of 42×10^{-6} μCi ^{89}Sr per μCi ^{89}Kr at maximum ingrowth (after 100 minutes). At the average gaseous fission product release rate of 12,500 μCi/sec during operating periods in 1968, and a 1.2 percent ^{89}Kr content relative to the nominal gaseous fission product release at the stack (see Table B.3), ingrowth added ^{89}Sr at the rate of $42 \times 10^{-6} \times 12{,}500 \times 0.012 = 6.3 \times 10^{-3}$ μCi/sec to the 1.0×10^{-3} μCi/sec measured at the stack. Thus, the virtual release rate to air increases with travel time from the value measured at the stack (1.0×10^{-3} μCi/sec) to the total ingrowth value (7.3×10^{-3} μCi/sec) at 100 minutes and longer.

The longer-lived radionuclides, ^{60}Co, ^{90}Sr, and ^{137}Cs can be used as indicators of long-term accumulation. Measurement of ^{137}Cs was emphasized because, of the three, it was released at highest average concentration (see Table 4.6) from the stack, and also is formed in air by decaying ^{137}Xe. The activity of ^{137}Cs reaches 0.25×10^{-6} μCi per μCi of ^{137}Xe after 25 minutes, and 2.8 percent of the nominal gaseous fission product release rate at the stack is ^{137}Xe (Appendix B.3). Hence, ingrowth added ^{137}Cs at the rate of $0.25 \times 10^{-6} \times 0.028 \times 12{,}500 = 88 \times 10^{-6}$ μCi/sec to the average rate at the stack of 35×10^{-6} μCi/sec.

The results of this study are summarized by sample types in Sections 7.3 to 7.8. Only in a few instances—^{131}I in three cattle thyroids (Section 7.7), ^{89}Sr in one snow sample (Section 7.3), ^{137}Cs in one sample of field corn (Section 7.5)—may the detected radioactivity be attributable to stack effluent at Dresden. All other measurements are reported to describe analytical techniques (Section 7.6, for ^{131}I in milk) and to report the absence, at the indicated levels of detectability, of radionuclides attributable to Dresden.

Other information on radionuclides in the environment of Dresden is available in the report of Dresden's contractor for environmental surveillance.(1) The contractor's program at the time of this study included the following:

air filter gross beta and ^{131}I analysis

milk ^{131}I, ^{137}Cs, ^{89}Sr, and ^{90}Sr analysis

well water gross beta analysis

rainwater gross beta analysis

surface water gross beta, ^{3}H, ^{89}Sr, and ^{90}Sr analysis

vegetation gross beta, gross gamma, ^{89}Sr and ^{90}Sr analysis

soil gross beta, gamma-ray spectral, ^{89}Sr and ^{90}Sr analysis

external radiation exposure rates

In addition, the Illinois Department of Public Health has analyzed surface and well-water samples for gross alpha and beta activity;(2) the Argonne National Laboratory has analyzed water and silt samples in the DuPage River at Channahon and in the Illinois River at Dresden Dam and Morris for gross alpha and beta activity;(3) the Federal Water Pollution Control Administration (FWPCA) analyzes Illinois River water at Morris for ^{90}Sr and gross alpha and beta activity; and the Public Health Service (PHS) analyzes milk at Chicago for ^{89}Sr, ^{90}Sr, ^{131}I, ^{137}Cs, and ^{140}Ba, and surface air and precipitation at Springfield, Illinois, for gross beta activity. The FWPCA and PHS data are reported at regular intervals in *Radiological Health Data and Reports*.

7.2 Estimation of Radioactivity Concentrations

7.2.1 *Precipitation*. Deposition by washout, W (in pCi/m^2), for ^{89}Sr in rain and snow was estimated by the following equation, derived from equation 5.6 of Slade:(4)

$$W = \frac{Q'_o \, L \, T \exp(-L x/\bar{u})}{\theta \, \bar{u} \, x} \qquad (7.1)$$

where Q'_o: virtual release rate at stack, pCi/sec

L : washout coefficient, sec^{-1}

T : duration of washout, sec

θ : sector width, radians

$\bar{u}$: wind speed at release height, m/sec

x : distance from stack to sampling point, m

Values of Q'_o, T, θ, $\bar{u}$, x, and the resulting W are given in Appendix D.1. The virtual release rate for ^{89}Sr was the average release rate at the stack from Table 4.6, plus a value based on ^{89}Kr decay that increased with the travel time of the plume as indicated in Section 7.1.2. The value of L for ^{89}Sr was estimated to be 1×10^{-5} sec^{-1}.(5) Deposition values were converted to concentrations in water by dividing by the mm of rainfall, and by the cm of snowfall.

7.2.2 *Deposition.* To estimate dry deposition of ^{131}I on pasture grass for uptake by dairy cows and beef cattle, and the concentration of ^{89}Sr on soil, leafy vegetables, corn husks, and grass, equation 5.44 of Slade(4) was integrated with respect to the crosswind direction and then distributed across an appropriate sector width, generally 10°. The equation was generalized to:

$$D = \frac{(2/\pi)^{1/2}}{\theta} \cdot \frac{v_d Q'_x T}{x \sigma_z \bar{u}} \cdot \exp\left[-h^2/2\sigma_z^2\right] \qquad (7.2)$$

where:

D = deposition, pCi/m^2

v_d = deposition velocity, m/sec

Q'_x = depletion-corrected release rate at point of interest, pCi/sec

T = total duration of deposition, sec

θ = sector width, radians

x = distance to point of interest, meters

σ_z = standard deviation of vertical concentration distribution, meters

$\bar{u}$ = release height wind speed, m/sec

h = effective release height, meters

The deposition velocity on pasture grass and other vegetation was taken to be 3×10^{-3} m/sec(5) for ^{89}Sr and 1×10^{-2} m/sec(6) for ^{131}I. To compute the radionuclide concentration in grass, it was assumed that deposited activity was completely retained, that the average grass density was 0.33 kg dry weight/m^2,(7) and that ash constituted 7 percent of dry weight.(8) Concentrations in cabbage were based on a published ratio of concentration in cabbage hearts to ground contamination.(9) For corn husks, 10 percent retention of deposition and a crop yield of 1 bushel of whole ears per square meter were assumed.

Dry deposition at a distance--for the background samples--was simplified by computing average values over the entire time interval of interest for stable and neutral conditions. Deposition during unstable conditions and rainstorms was judged to be relatively insignificant during the periods of interest. Based on the derivation in Appendix D.2, the simplified equations at locations 22 and 26 km NNE were:

$$D_{\text{stable}} = \frac{0.19T}{7.93 \times 10^4} \qquad (7.3)$$

$$D_{\text{neutral}} = \frac{0.034T}{4.68 \times 10^4} \qquad (7.4)$$

D = deposition, pCi/m^2

T = interval duration, sec.

Deposition during rainfall near Dresden was computed according to equation 7.1. Concentrations in vegetation were derived from washout with the same factors that were used for dry deposition. The value of L for ^{131}I was selected from Slade's Figure 5.12.(4)

Examples of deposition estimates for ^{131}I and ^{89}Sr are given in Appendices D.3 to D.9. The deposited radionuclide was summed for the entire period that contributed toward the radioactivity at the time of measurement, as shown in Appendices D.7 to D.9, correcting for the decrease of radionuclide concentration through decay and transfer to other media. For estimating ^{137}Cs concentrations in soil, the following were assumed: an average virtual release rate at the stack of 72 pCi/sec (which includes partial ingrowth from the ^{137}Xe parent), the same deposition velocity and washout coefficient as for ^{89}Sr, complete retention in soil (except for radioactive decay) and 50 percent retention in the top 1 cm, and ^{137}Cs accumulating over the 7-year operating period, adjusted for 20 percent down-time.

7.2.3 *Uptake in cattle thyroids and transfer to cows' milk.* From the estimated deposition of ^{131}I during 20 days before milk collection, levels of ^{131}I in milk were predicted in Appendices D.11 and D.12 from the graph by Garner and Russell$^{(10)}$ for which values are shown in Appendix D.10. The daily intake of ^{131}I was related to levels in milk at specified times by correcting for decay of ^{131}I on grass ($T_{1/2}$ = 5 days), assuming that a cow effectively grazes 45 m^2 per day,$^{(7)}$ and summing ^{131}I doses on the dates of deposition.

The estimate of ^{131}I in the thyroids of three heifers that grazed for three weeks prior to slaughter near the Dresden stack was based on Garner and Russell's model (Appendix D.10). It was assumed that 20% of the daily intake of ^{131}I goes to the thyroid$^{(10)}$ and that ^{131}I turns over in the thyroid with a 7-day half life$^{(11)}$ (Appendix D.13).

7.3 Rain and Snow

7.3.1 *Samples and analyses.* Radionuclides released from the stack are transferred to soil and vegetation near Dresden by precipitation and dry deposition. A preliminary estimate suggested that collection of several hundred liters of rain water would be necessary to detect ^{89}Sr, ^{131}I, or ^{140}Ba from the Dresden stack at ground level. One set of 1-liter rain samples was collected, nevertheless, and analyzed for radiostrontium and radionuclides that emit gamma rays. Two snow samples were collected for the same purpose.

Rain water was taken from puddles and ditches within 3 days after the tropical storm of June 22–24, 1968, at the six locations near Dresden listed in Table 7.1; a rain collector at a dairy 22 km NNE of Dresden provided a background sample. Snow was collected four days after the snowfall of January 13–14, 1968, from the surface of areas approximately 1-m square, at the two locations shown in Figures 7.1 and 7.2. The area near the meteorological tower, 0.8 km SSW of the stack, was beneath the plume from Dresden during snowfall, but not in the period between the end of snowfall and sampling. The area at the intersection of highways US 6 and 66, 10 km NE of the stack, was upwind from Dresden during the snowfall. Upon melting, the downwind sample contained 3.5 liter, and the upwind sample, 5.0 liter.

The samples were evaporated to 40 ml and analyzed by gamma-ray spectrometry with 10 × 10 cm NaI(Tl) detectors. Aliquots were analyzed radiochemically for ^{89}Sr and ^{90}Sr. The rain samples were also analyzed radiochemically for ^{137}Cs, and the snow samples,

Table 7.1

^{89}Sr, ^{90}Sr, and ^{137}Cs in Rain Water, pCi/liter

#	Location	Collection date, 1968	Measured			Estimated
			^{137}Cs	^{90}Sr	^{89}Sr	^{89}Sr
1*	3.2 km W	June 25	< 0.2	2.7	< 1	2.0×10^{-4}
2	0.8 km S	25	< 0.2	2.0	< 1	0
3	0.8 km SSW	27	< 0.2	3.4	< 1	0
4	2.4 km ESE	26	< 0.2	5.6	< 1	5.2×10^{-4}
5	1.5 km N	26	< 0.2	1.3	< 1	7.2×10^{-4}
6	1.6 km NNE	27	0.4	3.6	< 1	0
7	22 km NNE	25	< 0.2	0.8	< 1	0

*Locations of #2 to 6 are shown in Figure 7.1, #1 and 7 are shown in Figure 7.2.

Note: < values are 3 σ counting errors.

for ^{3}H. Snow was analyzed for ^{60}Co by coincidence spectrometry.

7.3.2 *Results and discussion.* The rainwater samples do not appear to contain radionuclides from Dresden. Only ^{90}Sr was found in all samples (see Table 7,1); in the absence of ^{89}Sr, the ^{90}Sr is attributed to fallout. Concentrations of ^{89}Sr and ^{137}Cs were below the limits of detectability (except for one ^{137}Cs value), and no radionuclides (< 3 pCi/l) that emit gamma rays were found by spectrometry. Concentrations of ^{89}Sr from Dresden were estimated in Appendix D.1 to be three orders of magnitude below minimum detectable concentrations. The estimates are quite approximate, however, because values of all the predictive components, especially the washout coefficient, are so uncertain.

The downwind sample of snow near Dresden contained ^{89}Sr while the upwind sample did not (see Table 7.2), suggesting that the ^{89}Sr was released at the Dresden stack. The ^{89}Sr concentration of 2.3×10^{-2} pCi/liter, estimated in Appendix D.1, is lower by 400, but washout coefficients for snow are even more uncertain than for rain.(4) All other radionuclides are at similar concentrations in the two snow samples, and are therefore attributed to fallout from atmospheric nuclear weapon tests, especially the Chinese test on December 24, 1967. Higher-than-usual gross beta values in two samples collected early in January(12) by Dresden's contractor for environmental surveillance are also associated with this nuclear test.

Although the measured concentration of ^{89}Sr in snow is 300 times lower than the permissible concentration for drinking water of 3000 pCi/liter, and the less-than values of ^{89}Sr in rain water are even lower, additional measurements of ^{89}Sr in large precipitation samples would be desirable in view of the uncertain values of washout coefficients. Analysis of snow to check the detection of ^{89}Sr would be of special interest.

Table 7.2

Radionuclide Content of Snow, pCi/liter

Radionuclide	Near Meteorological Tower	Intersection US 6 & 66
^{3}H*	500 ± 200	600 ± 200
^{60}Co	< 1	< 1
^{89}Sr	10 ± 2	< 1
^{90}Sr	2.5 ± 0.1	2.0 ± 0.1
^{95}Zr	55	35
^{103}Ru	25	30
^{106}Ru	15 ± 10	< 10
^{131}I	10	10
^{137}Cs	< 10	15
^{140}Ba	40	25
^{141}Ce	35	25

*Analysis by SERHL.

Notes: All analytical 2 σ values are ± 5 pCi/liter except as shown; < values are 3 σ counting error.

Concentrations corrected for decay to Jan. 18, 1968.

7.4 Soil

7.4.1 *Samples and analyses.* Soil may accumulate radionuclides deposited in precipitation and from air, and transfer these radionuclides to food and vegetation; long-lived radionuclides such as ^{90}Sr and ^{137}Cs accumulate in soil over many years. Samples were collected at five locations within 0.9 to 3.2 km of Dresden, at the same location as the corn samples (see Figure 7.1), so that radionuclide concentrations in corn kernels and soil could be compared.

Soil was dried at 110°C, and samples of 400 cc—approximately 500 g—were analyzed by gamma-ray spectrometry with a 10×10 cm NaI(Tl) detector. Aliquots of 5 g were analyzed radiochemically for ^{89}Sr and ^{90}Sr, and other aliquots were analyzed for stable strontium and calcium.

7.4.2 *Results and discussion.* Gamma-ray spectral analysis showed ^{137}Cs (see Table 7.3) and the naturally occurring radionuclides ^{40}K, ^{232}Th, and ^{226}Ra. Concentrations of ^{90}Sr and ^{137}Cs were each approximately 0.3 pCi/g dried soil. No

Table 7.3

Concentration of Radionuclides and Stable Ions in Dresden Soil, August 21, 1968

Sample Location	Sample wt., g	Stable Ions mg/g dry soil			Measured Radionuclides, pCi/g dry soil			Estimated ^{137}Cs, pCi/g dry soil
		K	Ca	Sr	^{89}Sr	^{90}Sr	^{137}Cs	
#1 - 1.7 km NE	470	13	3.3	0.015	< 2	0.3 ± 0.1	0.40 ± 0.10	2×10^{-5}
#2 - 1.6 km N	470	13	6.5	0.020	< 2	0.3 ± 0.1	0.30 ± 0.08	2×10^{-5}
#3 - 1.7 km E	540	11	3.3	0.011	< 2	< 0.1	0.29 ± 0.05	2×10^{-5}
#4 - 3.2 km W	480	13	4.8	0.013	< 2	0.3 ± 0.1	0.29 ± 0.07	6×10^{-6}
#5 - 0.9 km S	300	10	3.5	0.011	< 2	0.2 ± 0.1	0.40 ± 0.10	2×10^{-5}

Notes: 1. ± values are based on 1 σ counting error for sample and < values are based on 3 σ estimated counting error in background, except that uncertainty in ^{137}Cs measurements is estimated to be ± 25%.

2. Estimated concentrations of ^{90}Sr are less than 1 percent of estimated ^{137}Cs values at every location.

^{89}Sr (< 2 pCi/g) was found. Dresden's contractor for surveillance reported ^{90}Sr concentrations of 1.8 and 1.6 pCi/g, and ^{137}Cs concentrations of 1.9 and 1.8 pCi/g in June, 1968.(1) Estimated ^{137}Cs concentrations, based on accumulation over 7 years as described in Section 7.2.2, were lower than measured values by four orders of magnitude, suggesting that the measured concentrations of the two radionuclides are not attributable to routine releases from the Dresden stack.

7.5 Food and Feed

7.5.1 *Samples and analyses.* Foods are a potential vector for the transfer of radionuclides to humans from both soil and air, but only a few private vegetable gardens grow food near Dresden. A bushel basket of leafy cabbage plus a few heads of leaf lettuce were obtained on June 26, 1968, from two gardens at Channahon, 3.8 and 5.4 km NE of Dresden (see Figure 7.1).* As a background sample of leafy vegetables, only mustard greens, purchased at a large truck farm near Plainfield, 26 km NNE of Dresden, were available.

A bushel basket of field corn was collected on August 21, 1968, at each of the five locations near Dresden shown in Figure 7.1; a bushel of sweet corn grown near Gibson City, Ill., 110 km S of Dresden, was obtained on August 22 for background measurements.** Grass was collected at two dairies, on the pastures used for grazing by the cows whose milk was analyzed (Section 7.6). The grass was cut on June 26, during the period of milk collection.

The leafy vegetables, grass, corn kernels and corn husks were each analyzed by gamma-ray spectrometry with the 10 x 10 cm NaI(Tl) detector. The samples were then ashed and reanalyzed by gamma-ray spectrometry. Aliquots were analyzed for ^{89}Sr, ^{90}Sr, stable strontium, and calcium.†

7.5.2 *Results and discussion.* The leafy vegetables and grass contained 2 to 6 pCi ^{90}Sr per gram ash and no detectable ^{89}Sr, as shown in Table 7.4. The gamma-ray spectra showed the usual longer-lived fission-products, including $^{141/144}$Ce, $^{103/106}$Ru, ^{137}Cs, and ^{95}Zr + ^{95}Nb, that were in fallout from nuclear tests at the time of sampling. The Dresden contractor, however, reported detectable concentrations

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*We thank Mr. A. McDonald and Mrs. Hulbert for providing these samples.

**We thank Messrs. Dirker, Vandborg, and Henker, Stokely-Van Camp, Inc., and the Commonwealth Edison Company for providing the corn.

†We thank Mrs. G. L. Rehnberg and Mrs. A. Strong, SERHL, for analyzing the leafy vegetables and corn.

Table 7.4

Radiostrontium Concentration in Vegetables and Grass, June 26, 1968

	Cabbage & Lettuce, 3.8 and 5.4 km NE		Mustard Greens, 26 km NNE		Grass, 3.4 km W		Grass, 22 km NNE	
	measured	estimated	measured	estimated	measured	estimated	measured	estimated
Wet weight, kg	3.42		7.35		---		---	
% Dry weight	11.1		6.8		---		---	
% Ash weight	4.29		1.12		---		---	
^{89}Sr, pCi/g ash	< 0.2	1.5×10^{-6}	< 0.2	8.4×10^{-6}	< 1.0	4.1×10^{-2}	< 1.3	1.9×10^{-2}
^{90}Sr, pCi/g ash	2.2 ± 0.2	---	1.8 ± 0.2	---	5.9 ± 0.2	---	2.3 ± 0.2	---
Ca, mg/g ash	62	---	93	---	44	---	25	---
Sr, μg/g ash	140	---	140	---	140	---	310	---

Notes: 1. ^{90}Sr values are ± 2 σ, based on estimated counting error; less-than values for ^{89}Sr are based on 3 σ estimated counting error.

2. Estimated ^{89}Sr values from Appendices D.7, D.8, and D.9.

of ^{89}Sr (1.6 to 1.9 pCi/g ash) in three vegetation samples collected in June; his strontium-90 concentrations were 3.5 to 3.6 pCi/g ash, similar to values reported here.(1) The radionuclides are attributed to fallout because the gamma-ray spectra were similar in the near-by and distant samples of grass and leafy vegetables, and because ^{89}Sr concentrations were lower than ^{90}Sr concentrations. The concentration of ^{89}Sr estimated from stack releases at Dresden to be in the nearby grass is approximately 20-fold lower than could be measured. Because the estimated values for the leafy vegetables are based on cabbage hearts, a more appropriate value may lie between the given estimates and those for grass. The results of the analyses of leafy vegetables should be checked by measurements of washed samples because the observed ash weight in this sample is unusually high. It is possible that the ash contained some soil.

Concentrations of ^{90}Sr and ^{137}Cs also were of the order of 1 pCi/g ash in corn husks (Table 7.5). Strontium-89 was not detected. Corn husks, like leafy vegetables and grass, would be expected to accumulate most radioactivity from deposition of airborne particles.

The corn kernels, which are relatively sheltered from airborne radionuclides, contained undetectably low levels of ^{89}Sr and ^{90}Sr; concentrations of ^{137}Cs were, with one exception, between 0.2 and 0.8 pCi/g ash (see Table 7.5). Gamma-ray spectra indicate only ^{137}Cs and naturally occurring ^{40}K (see Figure 7.3). The highest concentration of ^{137}Cs was 4.3 pCi/g ash in the corn grown near the meteorological tower, 0.9 km S of Dresden. The field corn is used for animal feed, not for human consumption. It would be of interest to find the reason for the elevated level of ^{137}Cs, especially since the ^{137}Cs concentration was not correspondingly higher in the soil sample.

7.6 Milk

7.6.1 *Samples.* Milk is a major potential vector for the transfer of radionuclides to the population. If dairies are located near nuclear power stations, milk samples are usually analyzed as part of the environ-

Table 7.5

Concentrations of Radionuclides and Stable Ions in Corn Kernels and Husks, August 21, 1968

Location	Wet wt. kg.	% Dry wt.	% Ash wt.	mg/g ash K	mg/g ash Ca	mg/g ash Sr	pCi/g ash ^{89}Sr	pCi/g ash ^{90}Sr	pCi/g ash ^{137}Cs
Kernels									
#1 - 1.7 km NE	4.21	46.9	0.87	350	1.2	0.009	< 1	< 0.1	0.2 ± 0.1
#2 - 1.6 km N	4.40	30.6	0.86	410	1.6	0.012	< 1	< 0.1	0.4 ± 0.1
#3 - 1.7 km E	3.94	35.1	1.02	380	1.5	0.014	< 1	< 0.1	0.8 ± 0.2
#4 - 3.2 km W	3.48	43.5	0.86	350	1.7	0.015	< 1	< 0.1	0.8 ± 0.2
#5 - 0.9 km S	2.44	30.6	0.81	380	2.6	0.017	< 1	< 0.1	4.3 ± 0.2
#6 - 110 km S	3.82	37.1	0.92	390	1.7	0.014	< 1	< 0.1	0.7 ± 0.2
Husks									
#1 - 1.7 km NE	2.22	27.7	1.66	240	18.7	0.036	< 1	0.7 ± 0.3	0.4 ± 0.2
#6 - 110 km S	1.72	20.5	1.00	290	19.2	0.050	< 1	0.6 ± 0.3	1.1 ± 0.2

± value based on 1 σ estimated counting error for particular sample.

< values based on 3 σ estimated counting error for background sample.

Standard deviation of duplicate analyses was ± 0.3 mg calcium/g ash, ± 10 percent for potassium, and ± 25 percent for stable strontium.

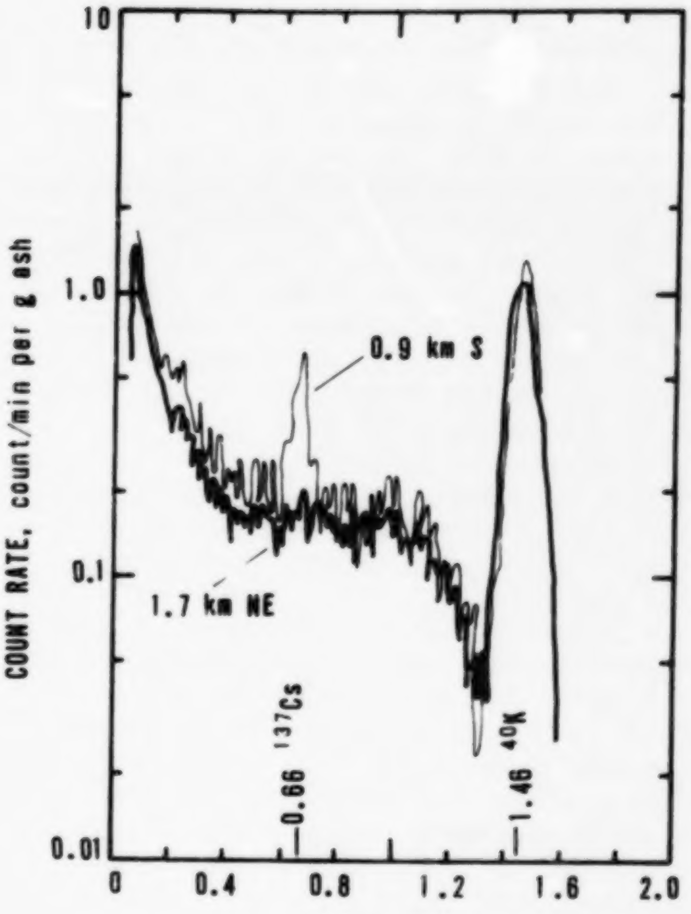

Figure 7.3. Gamma-ray spectra of corn kernels. Detector: 10 × 10 cm NaI(Tl) at SERHL. Sample: 11.3 g ash of corn collected 0.9 km S of Dresden and 21.7 g collected 1.7 km NE of Dresden on 8-21-68. Counts: 50 min. on 9-12-68 (bgd subtracted).

mental surveillance program. At Dresden, the contractor for environmental surveillance collects weekly composites of daily samples at two dairies located 10 and 22 km NNE of Dresden. He reports weekly average concentrations of ^{131}I and monthly averages of ^{89}Sr, ^{90}Sr, and ^{137}Cs.(1) Any significantly higher values at the nearby dairy would suggest that radionuclides from Dresden have entered the milk. During the second quarter of 1968, the ranges of reported values were as follows:(1)

^{131}I	0.8 - 3.8 pCi/liter
^{137}Cs	7.0 - 13.1
^{89}Sr	2.7 - 4.6
^{90}Sr	2.4 - 7.4

Concentrations at the nearer dairy were not significantly higher than at th- farther dairy, suggesting that these radionuclides originated in fallout from atmospheric nuclear weapon tests.

A preliminary estimate of radionuclide concentrations in milk at the nearer dairy, based on radionuclide release data in Table 4.5, indicated that ^{131}I could be detected with the greatest sensitivity among these radionuclides, but that its concentration would be below the limit of detectability of 0.1 pCi/liter. It was considered more probable that ^{131}I could be detected in milk of the dairy herd nearest Dresden, 3.4 km west (see Figure 7.2). The herd consists of 11 Holstein cows, and the milk is grade B—not bottled for sale. During the grazing season, however, the wind very infrequently blows from Dresden toward the pasture. After awaiting this occurrence throughout spring and summer of 1968, it appears that the recorded incidence of winds at 75° to 85° consists mostly of rotation through this angle for short periods.

Upon observing that the wind had been in the 75° to 85° sector for several hours on June 22, milk samples were collected at the dairy west of Dresden in the evenings of June 25 and the following two days.* Each consisted of 45 liters of milk produced by 5 cows selected at random. For comparison, 45-liter samples were taken on the first and third day at the farm 22 km NNE of Dresden that supplies weekly composites to Dresden's contractor for environmental surveillance.* These samples represented the combined milk of all cows (12-15 Holsteins). One sample at each location had been collected previously to test collection and analytical procedures.

7.6.2 *Analysis.* To concentrate ^{131}I, 22.5-liter portions of each sample were passed through 80 ml Dowex 1-x8 anion-exchange resin (Cl$^-$, 20-50 mesh) in a 3.1-cm-diameter column. The flow rate was initially 100 ml/min and decreased slowly with time. The resin was washed with water, transferred to a Petri dish (9.0-cm dia.

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*We thank Messrs. A. Dirker and H. Dhuse for the milk samples.

x 1.5 cm high), and counted for 300 to 1000 minutes with either a 10 x 10 cm NaI(Tl) detector plus multichannel analyzer, or two such detectors facing each other within an annular anticoincidence shield. A 100-keV range at the 364-keV characteristic photopeak was used to compute the amount of ^{131}I in the sample.

Recovery of ^{131}I by this procedure had been found to be 90 ± 5 percent by tracer tests with *in vivo* spiked cows' milk and by comparison with gamma spectral analysis of milk that contained ^{131}I from fallout at the Nevada test site.(13) The minimum detectable concentration, based on the 3-sigma statistical deviation at a background of 30 counts per minute and a counting efficiency for ^{131}I of either 30 percent (sum/anticoincidence) or 16.5 percent (single detector) was 0.07 and 0.13 pCi/liter, respectively. Examination of the reproducibility of background measurements and of ^{131}I samples in the range of 1 to 10 pCi confirmed a minimum detectable value of 0.1 pCi/liter. In late June and early July, however, high background values in the counters increased the minimum detectable levels (3 sigma of deviation of background values for 300 to 1000 minutes) to 0.3 and 0.5 pCi/liter, respectively, at the time of counting. Correction for radioactive decay from the time of sampling increased these minimum detectable values.

7.6.3 *Results and discussion.* No ^{131}I was found, as shown by the ''less-than'' values in Table 7.6, suggesting that neither fallout from atmospheric testing of nuclear devices nor releases from Dresden caused ^{131}I concentrations in milk to reach 0.6 pCi/liter. The values for the herd 22 km NNE of Dresden disagree with a concentration of 1.6 ± 0.4 pCi/liter in Table 3 of the report by Dresden's contractor.(1) The ^{131}I detected in the contractor's weekly composite may have originated in samples from the 5 other days, but a more probable cause is systematic error in analysis by a small but constant amount.

The concentrations predicted in Appendices D.11 and D.12 from stack releases support the measured ''less-than'' values, as shown in Table 7.6. Estimated concentrations in the milk of cows grazing 3.4 km west of Dresden for ^{131}I released from the stack were approximately 10-fold less than could be measured, and concentrations in the milk of cows grazing 22 km NNE were 50-fold lower. The estimated values are quite approximate, in that variations by factors of at least two may be expected in each of the following: release rate, dilu-

Table 7.6

^{131}I Concentration in Raw Milk, pCi/liter

Date, 1968	3.4 km W		22 km NNE	
	measured	estimated	measured	estimated
May 7	< 0.30 < 0.14	0*	< 0.17 < 0.17	0*
June 25	< 0.60 < 0.71	0.047	< 0.67 < 0.60	0.013
June 26	< 0.71 < 0.71	0.075	--- ---	---
June 27	< 0.67 < 0.71	0.090	< 0.62 < 0.71	0.013

Notes: 1. Directions and distances are from Dresden stack.
2. ''Less-than'' values are 3-sigma for measured mean detector backgrounds.
3. Duplicate samples were analyzed for 4 evening milkings at nearer farm, and for 3 evening milkings at farther farm; values are at time of milking.

*Dresden reactor was shut down for refueling.

tion in air, deposition velocity or washout coefficient, transfer from grass to cow, and metabolic transfer to milk.

It would be desirable to repeat these measurements at the nearer pasture under circumstances that might yield definite ^{131}I values. For example, improved stability in detector background would reduce minimum detectable concentrations to approximately 0.1 pCi/liter; and the five-fold higher releases of ^{131}I that have been measured (see Table 4.5) would lead to ^{131}I concentrations of 0.3 pCi/liter under similar meteorological conditions. On the other hand, occasions for sampling at detectable concentrations are infrequent because prevailing winds are not to the west, and the farm has been sold, so that dairy operations at this location will probably cease.

7.7 Cattle Thyroids

7.7.1 *Samples.* Cattle thyroids are not eaten (to the best of our knowledge) and therefore are not a vector in the transfer of ^{131}I to man. They are sensitive indicators of ^{131}I on pasture, and potentially in milk, however: 12 pCi ^{131}I/g of thyroid has been taken to be equivalent to 1 pCi ^{131}I/liter of milk.(14) Small herds of beef cattle were observed to be on pasture in several directions from Dresden within a few kilometers.

Use of cattle thyroids as indicators for ^{131}I can only be recommended under defined conditions. By common practice, beef cattle are removed from pasture and given stored feed for several weeks before slaughter, so that any ^{131}I from grazing would have decayed appreciably at the time of slaughter. Metabolic and dietary variations significantly affect the uptake of ^{131}I by the thyroid, and may reduce it drastically.(15) Moreover, the relative concentrations of ^{131}I in milk and thyroid are not constant, but depend on the time interval between uptake by the cow and measurement.

At our request, three heifers were kept on pasture 2.3 km east of Dresden (see Figure 7.1) from June 16 until they were shipped on July 7 for sale and slaughter.* The three heifers were "back-tagged" to identify them at slaughter. Their thyroids were removed, placed in formaldehyde and mailed at Green Bay, Wisconsin on July 10.** Thyroids of five steer from more distant locations were obtained on July 29 at Morris, Ill.† Although intended as "background" samples, they were not, because of the difference in date of slaughter and the stored feed consumed by these cattle before slaughter.

7.7.2 *Analysis.* The thyroids were measured for ^{131}I content by placing them in Petri dishes on the NaI(Tl) detector or within the 2-detector/annulus system, in the same way that ^{131}I in milk had been analyzed (Section 7.6.2). Spectra were accumulated for 300 to 1,000 minutes, and ^{131}I was determined from the count rate of the characteristic 364-keV gamma ray. The minimum detectable ^{131}I activity at the time of counting (3 sigma of counting error) was 3 pCi; computed minimum detectable levels at sampling were higher because of ^{131}I decay.

7.7.3 *Results and discussion.* As shown in Table 7.7, thyroids from the three heifers that had grazed 2.3 km east of Dresden contained measurable amounts of ^{131}I, while the other thyroids did not. The magnitude of the ^{131}I peak in the former thyroids is indicated in Figure 7.4. The estimated value for cattle grazing 2.3 km east of Dresden, computed in Appendix D.13, agrees with the average of the three measured values. The estimation is subject to all the uncertainties discussed in Section 7.6.3, and the agreement therefore is appreciably better than expected. At the 12/1 ratio, the indicated ^{131}I concentration in milk, if milk cows had grazed on the same pasture, would be 0.04 pCi/liter.

The excellent agreement between measured values and the estimate based on ^{131}I release at the Dresden stack suggests that

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*We thank Mr. M. McDonald for making these heifers available to the study.

**We thank Dr. LeVan, DVM, Meat Inspection Program, USDA, for providing these samples.

†We thank Mr. Floyd Schaible, Circle S Foods, for providing these samples and Dr. R. Conner, DVM, for identifying the thyroids at slaughter.

Table 7.7

^{131}I Content of Cattle Thyroids

Sample	Location	Date of slaughter	Thyroid wt., g	^{131}I Content at slaughter, pCi
2-yr old heifers	2.3 km E	July 10, 1968	73	39 ± 6
			78	23 ± 5
			58	30 ± 6
Average				31
Estimated				28
2-yr old steer	21 km ENE	July 29, 1968	27	< 9
	26 km W		26	< 5
	38 km SW		69	< 7
	5.6 km NW		24	< 5
	11 km N		27	< 6

Notes: 1. Directions and distances are from stack.
2. ± values are at 1-sigma level for counting error.
3. ''Less-than'' values are 3-sigma for measured mean.
4. Values are at time of slaughter.

Dresden was the source of the ^{131}I. In the absence of the ''background'' samples, however, one can not be certain. If the ^{131}I analyses in milk by the Dresden contractor (see Section 7.6.3) are correct, for example, the ^{131}I in the thyroids may have originated in fallout. Routine analyses of cattle thyroids collected throughout the United States in April, May, and June show only a single value above the ''barely detectable level'',(16) but these analyses are ten times less sensitive than in this study.

Analysis of cattle thyroids for ^{131}I content appears to be a relatively sensitive and convenient procedure for relating stack releases to the amount deposited on the ground and expected in milk. Simultaneous measurement of thyroids obtained at a greater distance is necessary, however, and the cattle diet must be controlled. Radionuclide analyses of the meat from beef cattle that had been on pasture near nuclear power stations would also be desirable.

7.8 Radionuclides in Wildlife

7.8.1 *Samples and analyses.* Animals in the neighborhood of Dresden that are hunted include squirrel, rabbit, deer, opossum, raccoon, muskrat, beaver, weasel, skunk, fox, duck, pheasant, quail, and crow. Hunting for all except fox and crow is restricted to short seasons. Rabbit, quail, and deer (with arrow) may be hunted in the Des Plaines Wildlife Conservation Area in the triangle between the Des Plaines and Kankakee Rivers, to the east of Dresden. Hunting is forbidden in the McKinley (Channahon) State Forest Preserve, north of the Des Plaines River and east of Dresden.(17)

To begin evaluating the radionuclide content of wildlife in the area, 3 rabbits and 2 deer were collected near Dresden, and 2 deer for background values at distances of 27 and 40 km.* The rabbits were shot in the McKinley State Forest Preserve, at point R in Figure 7.1. The nearby deer were obtained 7 km E and 10 km SE from Dresden (see Figure 7.2), after they had been struck and killed by automobiles. All samples were wrapped in plastic bags and preserved in dry ice until dissection.

*We thank Herman Heir, Jr., District Game Biologist, State of Illinois, and Ray Heintz, Game Warden, Will County, for collecting the deer and rabbit samples.

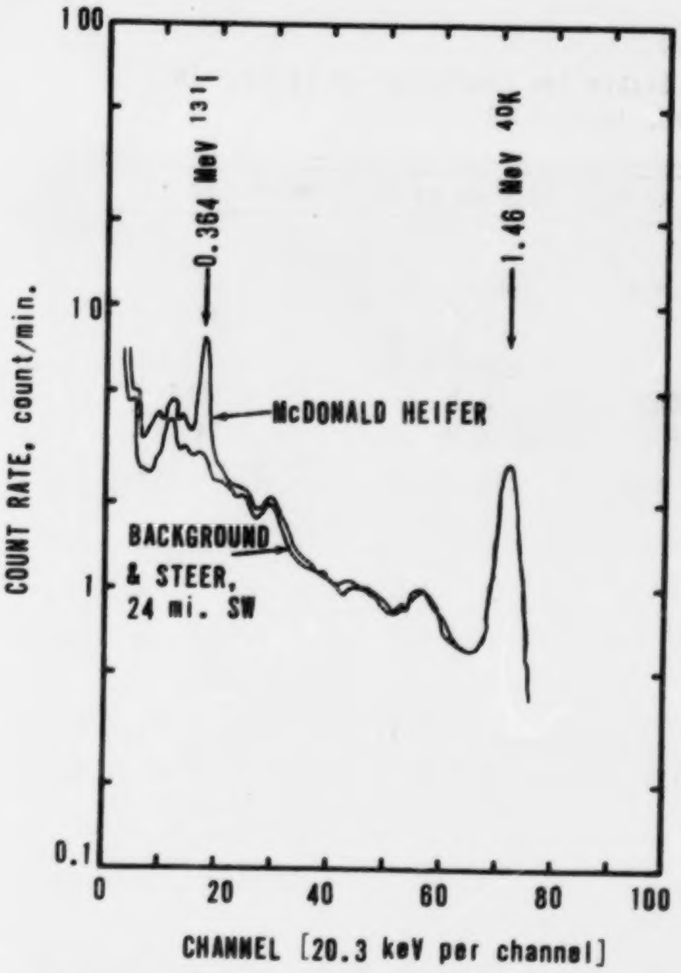

Figure 7.4. Gamma-ray spectra of cattle thyroids. Detector: Two 10 x 10 cm NaI(Tl) in anticoincidence shield. Sample: 73-g heifer and 69-g steer thyroids. Count: Heifer – 600 min. on July 12. Steer – 1000 min. on Aug. 9–10.

Separate analyses were performed for thyroids, liver plus kidney, muscle, and bone. In addition, rabbit skins and carcasses and deer rumen contents were analyzed. Muscle, liver plus kidney, and rumen content were ashed at 450°C; bone was ashed at 600°C; and thyroids were not ashed. All samples were analyzed by gamma-ray spectrometry with a 10 x 10 cm NaI(Tl) detector. Radiostrontium was determined radiochemically in bone (femurs) and rumen content.

7.8.2 *Results and discussion.* As shown in Table 7.8, radionuclide concentrations in deer rumen, muscle, and bone were too variable for detecting any distinction between the nearby samples (#3 and 4) and the relative distant ones (#1 and 2). The rumen contained ^{90}Sr, ^{95}Zr + ^{95}Nb, and ^{137}Cs; bone contained ^{90}Sr; and muscle contained ^{137}Cs. All radionuclides are attributable to fallout from atmospheric nuclear weapon tests. Gamma-ray spectra of the rumen contents showed also naturally occurring ^{40}K and the progeny of ^{226}Ra and ^{232}Th, and spectra of muscle also showed ^{40}K. No radiocobalt was found in liver plus kidney, no ^{89}Sr ($<$ 500 pCi/kg) in bone and rumen, and no ^{131}I in thyroids.

Concentrations of ^{137}Cs in the muscle of white-tailed deer range from 100 pCi/kg to more than 100,000 pCi/kg,(18,19) hence the average ^{137}Cs content of 67 pCi/kg in the analyzed deer is among the lowest values. Highest concentrations are observed in late winter at specific locations;(18-20) at these locations, soils of low-mineral content may favor growth of grasses with high fractional uptakes of minerals from the soil, and these grasses may provide the high-radiocesium diet for deer. The accumulation factor, pCi ^{137}Cs/kg muscle per pCi ^{137}Cs/kg rumen content, ranged from 0.4 to 2.6 in the four samples, compared to reported values of 3.3(18) and 0.68.(19)

The ^{90}Sr/calcium ratios of the deer bones ranged from 11 to 38 pCi/g, with an average value of 23 pCi/g. Similar values were observed in Colorado mule deer,(21) and higher values, at the Savannah River plant site(19) and in Alaska caribou.(22,23) The OR$_{bone/diet}$—the Sr/Ca ratio in bone divided by that in the diet (taking the rumen content to be the diet)—averaged 0.16 for ^{90}Sr relative to calcium, and 0.077 for stable strontium relative to calcium.

Gamma-ray spectral analyses of the rabbit tissues showed only ^{40}K in the skins and ^{137}Cs and ^{40}K in the carcasses. The concentration of ^{137}Cs (Table 7.9) was approximately 110 pCi/kg muscle (27 pCi/g potassium), and of ^{90}Sr, 1,400 pCi/kg bone (26 pCi/g calcium, if the calcium content in bone is assumed to be the same in the three rabbits). No radiocobalt was found in liver plus kidney, no ^{89}Sr in bone, and no ^{131}I in thyroids. Thus, although samples collected at a distance are necessary to

Table 7.8

Radionuclide Concentration (pCi/kg)* and Stable Ion Concentration (g/kg)* in Deer Samples Collected June, 1969

Substance	#1 - 27 km SE	#2 - 40 km NE	#3 - 10 km SE	#4 - 7 km ESE
Rumen content				
^{137}Cs	30 ± 3[†]	100 ± 8	140 ± 9	34 ± 2
K	2.0	2.8	4.6	1.02
^{90}Sr	48 ± 5	144 ± 9	320 ± 16	65 ± 4
Sr	0.0033	0.0062	0.0073	0.0018
Ca	0.73	1.08	1.56	0.34
^{95}Zr + ^{95}Nb	5 ± 2	13 ± 2	280 ± 20	16 ± 2
Muscle				
^{137}Cs	49 ± 5	69 ± 5	61 ± 6	90 ± 7
K	4.03	4.13	2.77	3.76
Bone				
^{90}Sr	1230 ± 90	2060 ± 100	1600 ± 70	3170 ± 150
Sr	0.039	0.034	0.042	0.029
Ca	111	80	101	84

*kg wet weight.

[†] ± values are 2 σ counting error.

evaluate the background from fallout, no radionuclides were found that would be clearly attributable to Dresden.

Analysis of deer and rabbits is desirable to evaluate their contribution to the dietary intake of radionuclides by humans, and may also be useful for monitoring environmental vegetation. The latter has been suggested with regard to ^{90}Sr in bones of jack rabbits at the National Reactor Test Site, Idaho, where values of approximately 15 pCi/g calcium were observed off-site between 1956 and 1960.(24) Most of the retention of ^{90}Sr in rabbit bone was found to occur during the first year of life. The observed ratio from diet to bone, $OR_{bone/diet}$, for strontium relative to calcium is 0.12 in the rabbit, compared to approximately 0.25 in man, and the average stable strontium/calcium ratio in

Table 7.9

Radionuclide (pCi/kg)* and Stable Ion (g/kg)* Concentrations in Tissue of Rabbits Shot on August 21, 1968

Tissue	Substance	Rabbit #1	Rabbit #2	Rabbit #3
Muscle	^{137}Cs	114 ± 12[**]	114 ± 10	91 ± 10
	K	4.5	3.5	3.8
Bone	^{90}Sr	1270 ± 70	1560 ± 90	1330 ± 70
	Sr	0.022	0.025	0.023
	Ca	54		

*kg wet weight.

[**] ± values are 2 σ counting error.

rabbit bone is 0.52 mg/g.(25) For ^{137}Cs, the rabbit body at equilibrium contains approximately 8 times its daily intake;(26) in man, the ratio of body burden to the daily intake is approximately 100.

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8. SUMMARY AND CONCLUSIONS

8.1 Radionuclides in Dresden Effluents

One of the purposes of this study was the identification and quantification of individual radionuclides in Dresden effluents. Radionuclides in liquids on site and in gaseous, airborne particulate, and liquid wastes were therefore measured during all five trips in the 9-month period of study; the results are presented in Sections 2, 3, and 4, and are summarized below. It should be noted, however, that only continuous measurements over extended periods can yield definitive values.

1. Average release rates of the measured noble gas fission products were:

4.4-hr	^{85m}Kr	3×10^{2} μCi/sec
10.7-yr	^{85}Kr	1×10^{-1}
76 -min	^{87}Kr	7×10^{2}
2.8-hr	^{88}Kr	5×10^{2}
2.3-d	^{133m}Xe	1×10^{1}
5.3-d	^{133}Xe	3×10^{2}
9.1-hr	^{135}Xe	8×10^{2}
17 -min	^{138}Xe	2×10^{3}

The release rates were measured in the delay line for off-gas from the condenser air ejectors. The average values were computed for the 1968 average fission-product noble gas release rate of 12,500 μCi/sec during 64 percent of the year. These values are generally in accord with measurements performed earlier by General Electric Co. staff.

2. Average release rates of the other measured radionuclides were:

Radio-nuclides	in stack effluent, μCi/sec	in liquid effluent, μCi/sec
^{3}H	6×10^{-3}	5×10^{-2}
^{58}Co	2×10^{-5}	2×10^{-2}
^{60}Co	2×10^{-5}	3×10^{-2}
^{89}Sr	7×10^{-4}	8×10^{-3}
^{90}Sr	3×10^{-6}	9×10^{-4}
^{131}I	6×10^{-4}	1×10^{-3}
^{134}Cs	$< 1 \times 10^{-5}$	2×10^{-3}
^{137}Cs	2×10^{-5}	6×10^{-3}
^{140}Ba	3×10^{-4}	5×10^{-3}
^{144}Ce	$< 3 \times 10^{-5}$	2×10^{-4}

The following should be noted concerning these values: (a) stack effluent values take into account operation during only 64 percent of the time in 1968; the liquid wastes that contained most of the radionuclides were released during less than one-half of the year; (b) for stack effluent, radioactive particles were measured on filters in the stack, ^{131}I was measured on charcoal cartridges in the stack, and ^{3}H was measured in the delay line for off-gas from the condenser air ejectors; (c) release rates in liquid effluent were computed from radionuclide concentrations measured in waste tanks before discharge; they can be converted to concentrations in water at the point of release by dividing by the flow rate in the coolant-water canal of 1×10^{7} ml/sec; (d) the ^{131}I release rate in the stack has been compensated for an 88-percent collection efficiency (see Section 4.3.3); (e) effective release rates of

^{89}Sr and ^{137}Cs are higher than measured because of formation by decay of ^{89}Kr-^{89}Rb and ^{137}Xe, respectively, in the environment (see Section 7.2.2); (f) concentrations of radiocobalt and radiocesium in water at the point of discharge from Dresden were considerably lower than indicated by the above release rates; it is probable that radiocobalt was mostly insoluble and that radiocesium was mostly retained on silt; (g) the measurements show considerable variations in concentrations and release rates.

3. The measured release rates are consistent with the average annual releases reported by Dresden Nuclear Power Station for the year 1968 of 0.189×10^{-7} μCi/ml of water and $12{,}500 \times 0.64 = 8{,}000$ μCi/sec in air. The sum of measured radionuclides in liquid effluent (see above) of 12×10^{-2} μCi/sec, divided by the flow rate of 1×10^{7} ml/sec, is 0.12×10^{-7} μCi/ml. The sum of measured noble gas fission products (see above) is 4,600 μCi/sec; the gases ^{83m}Kr, ^{89}Kr, ^{135m}Xe, ^{137}Xe, and ^{13}N would be expected to contribute additional activity amounting to several thousand μCi/sec.

4. The influence of plant operation on radionuclide release rates in effluent is indicated by comparing these release rates to fission production rates in the reactor core. The ratios of the fission product release rates shown above to estimated production rates (see Appendix B.1, adjusted for 64 percent operation 1968) are:

noble gas fission products		other fission products	
^{85m}Kr	1×10^{-6}	^{3}H	1×10^{-3}
^{85}Kr	5×10^{-5}	^{89}Sr	4×10^{-9}
^{87}Kr	6×10^{-7}	^{90}Sr	6×10^{-8}
^{88}Kr	7×10^{-7}	^{131}I	2×10^{-10}
^{133m}Xe	6×10^{-5}	^{137}Cs	4×10^{-7}
^{133}Xe	1×10^{-5}	^{140}Ba	4×10^{-10}
^{135}Xe	2×10^{-6}	^{144}Ce	4×10^{-10}
^{138}Xe	2×10^{-7}		

Thus, only small fractions of these fission products are released. The ratio for fission-produced tritium is actually lower than shown because most of the discharged tritium is probably produced by neutron activation of deuterium in reactor cooling water.

8.2 Radionuclides and Radiation from Dresden in the Environment

The main effort in the environmental aspect of this study was devoted to measuring radionuclides in the plume from the Dresden stack and the external radiation exposure from these radionuclides. The plume was detected as much as 18 km distant from the stack with large NaI(Tl) survey instruments. Within 1 to 2 km from the stack, radiation exposure rates were measured at the centerline of the plume under stable and neutral conditions with a tissue-equivalent ionization chamber and sensitive electrometer. Average exposure rates measured during three tests for 1/2- to 1-hour periods were 13, 24, and 40 μR/hr. Concentrations of ^{133}Xe, ^{135}Xe, and ^{138}Cs in ground-level air were measured at the same time as the radiation exposure rates (see Section 5.2.5). Estimated concentrations and exposure rates, based on measured release rates at the stack and diffusion calculations, were within a factor of two or better of the measured values.

Thermoluminescent dosimeters exposed during two 2-week periods at ten stations located between 1 and 4 km from Dresden showed an average background radiation exposure rate of 9 μR/hr. During reactor operation, TLD's at three locations indicated radiation exposure from the plume at average rates for a 2-week period of 2 to 3 μR/hr above the natural radiation background. The TLD values are uncertain, however, because their standard deviation is ± 1 μR/hr, and variation in the background could have introduced significant error.

Samples collected in the Dresden environment for analysis of individual radionuclides usually contained radionuclides such as ^{90}Sr and ^{137}Cs from fallout associated with atmospheric testing of nuclear devices, and naturally occurring radionuclides such as ^{40}K, ^{232}Th plus progeny, and ^{226}Ra plus progeny. To search for radionuclides attributable to Dresden, nearby samples were compared with more distant samples, and downstream or downwind samples with those collected upstream or upwind. In addition, measurement of ^{58}Co, ^{60}Co, ^{89}Sr, ^{131}I, and ^{134}Cs was emphasized, because these were discharged at Dresden while their concentrations in fallout were low or undetectable.

No radioactivity attributable to Dresden was found in the following samples:

rainwater (Section 7.3.2)
soil (Section 7.4.2)
cabbage (Section 7.5.2)
grass (Section 7.5.2)
corn husks (Section 7.5.2)
milk (Section 7.6.3)
deer (Section 7.8.2)
rabbit (Section 7.8.2)
surface water (Sections 6.2.2 and 3.3.7)
drinking water (Section 6.6.2)
fish (Section 6.3.2)

Estimated concentrations and measured less-than values suggest that a sensitivity of approximately 0.05 pCi/liter appears necessary to measure ^{131}I from Dresden in milk, and 0.001 pCi/liter to measure ^{89}Sr from Dresden in rainwater, for example.

Indications of radioactivity from Dresden were found in the following samples:

1. The average ^{131}I content of three thyroids of heifers was 31 pCi (0.45 pCi/g). These heifers had been placed on pasture 2.3 km east of Dresden for several weeks before slaughter specifically for testing cattle thyroid as a sensitive indicator of ^{131}I deposition on pasture grass.
2. Snow collected 0.9 km south of Dresden contained ^{89}Sr at a concentration of 10 pCi/liter, while snow collected 10 km NNE of Dresden contained no ^{89}Sr (< 1 pCi/liter). The area to the south of Dresden was downwind during the snowfall.
3. Kernels of field corn grown at the same location south of Dresden contained ^{137}Cs at a concentration of 4 pCi/g ash (at 0.8 percent ash weight), compared to 0.2 to 0.8 pCi/g ash in corn kernels from other locations.

On the basis of these measurements, exposure to the surrounding population through consumption of food and water from radionuclides released at Dresden was not measurable. External exposure from radioactive gases discharged from the Dresden stack was detectable, but it was only a small fraction of the natural radiation background over an extended period of time, and well within Federal Radiation Council guidance.

8.3 Monitoring Procedures

The following techniques were demonstrated in this study for monitoring radionuclide release and environmental transport:

1. analysis by gamma-ray spectrometer with Ge(Li) detectors of multiple radionuclides in samples of water from the station and in effluent before dilution;
2. radiochemical analysis of 35 radionuclides in the primary coolant;
3. collection of radioxenon from environmental air on charcoal for analysis by gamma-ray spectrometry;
4. collection of ^{138}Cs in environmental air on filters with high-volume samplers, and analysis by gamma-ray spectrometry;
5. use of portable NaI(Tl) survey meters as sensitive detectors of the plume from the Dresden stack;
6. use of muscle-equivalent ionization chamber and Shonka electrometer for quantifying the radiation exposure rate during brief periods within or beneath the plume;
7. use of TLD's for a 2-week period to quantify the average long-term radiation exposure rate from the plume;
8. collection of ionic radionuclides in water from 100-liter volumes on ion-exchange resins, and analysis by gamma-ray spectrometry and radiochemical separation;
9. collection of radioiodine from 22.5-liter volumes of milk on anion-exchange resin, and analysis by gamma-ray spectrometry;
10. measurement of ^{131}I in thyroids of cattle that had grazed under defined conditions near station;

11. collection and analysis of food samples, including vegetables, milk, fish, rabbit, and deer; and of drinking water;
12. use of measured release rates at the station, meteorological data, and transfer coefficients to estimate radionuclide concentrations in samples for comparison with measured or minimum detectable values.

Additional tests are required for several of the techniques; for example, no ^{131}I was detected in milk, and detection of external radiation with the TLD's was marginal.

8.4 Considerations in Developing Recommendations for Environmental Surveillance

The available guidance for radiological surveillance at nuclear power stations referred to in Section 1.1 and the experience from this study suggest a change from past emphasis at stations in the U.S. on monitoring gross radioactivity in effluent and extensively sampling the environment. The following approach is presented for consideration:

1. thorough monitoring of significant individual radionuclides in effluents;
2. specific studies of predicted critical nuclides, pathways, and populations by measuring transfer coefficients from plant to man in the environment;
3. direct measurement of radiation and radionuclides at the point of population exposure; and
4. periodic checking of potential concentration media for radionuclides in pathways from plant to man.

Many of the techniques for measuring individual radionuclides in effluents are available, some were demonstrated in this study (see Section 8.3), and others remain to be developed. Significant nuclides that should be measured include the critical radionuclides, the major radioactive constituents, and radionuclides that serve as indicators for more hazardous ones because they are easily measured. The list of radionuclides to be measured should be changed as it becomes possible to predict release rates for previously measured radionuclides and as interest becomes focussed on others. A well-designed program should make it possible to obtain more useful dosimetric information by monitoring individual radionuclides in effluents and computing the potential radiation exposure to man, than by the usual determination of ''less-than'' values in environmental samples.

If effluent monitoring is to be a major component of radiological surveillance, then accurate transfer coefficients from the point of release through the station environment to the population at risk must be developed. These can be obtained during the early years of station operation by undertaking specific studies, as demonstrated at Dresden in comparing the release rate of gaseous fission products at the stack with the radiation exposure rate beneath the plume in the environment. The studies can usually be performed within a few years of startup with radionuclides in effluents; alternatively, radioactive or stable tracers may be used before or soon after initial operation. As more nuclear stations begin operation and undertake these studies, the accumulation of information on the ecological cycling of radionuclides will decrease the need for further research, until only unusual waste management practices or environmental conditions will require such research.

Since control of exposure to man is of primary importance, it is necessary to monitor exposure directly. For these measurements, ''less-than'' values of external radiation exposure, radionuclide intake, or radionuclide body burden attributable to station operation are acceptable if the values are sufficiently below limits. After effluent monitoring and measurements of environmental transfer coefficients have provided quantitative data for predicting radiation exposure to the population, the frequency and intensity of monitoring radiation exposure should be adjusted to respond to the level of radionuclide releases.

Finally, some environmental measurements are desirable to maintain surveillance of potential concentrating media for radionuclides. The program of these measurements should be flexible in responding to changes in station operation, the environment and the population, to new information on uptake and transfer of radionuclides, and to unusual analytical results. As the knowledge of the environment increases, fewer measurements will be needed.

A more source-oriented program than is now customary is recommended because large nuclear power stations will be numerous, relatively close to population centers, and with little operating experience (except at considerably lower power levels) during the next few years. Over the long term, the program should provide more information at lower cost than many current environmental surveillance programs which appear to be exercises in measuring fallout radioactivity and the natural radiation background.

8.5 Suggested Future Studies

The following studies at nuclear power stations are suggested on the basis of the field trips to Dresden:

1. Incorporate and test the suggestions in Section 8.4 for a radiological surveillance program;
2. Test for extended periods several of the techniques that were demonstrated in this study, such as long-term measurements of external radiation exposure with dosimeters, calculation of long-term radiation exposure by short-term measurements, and use of concentration devices in analyzing radionuclides in water, milk, and air;
3. Relate radionuclide release rates to operating and waste-handling procedures to permit accurate estimation of release rates in future operations and for new stations;
4. Examine the effect of radioactive waste treatment on discharge practices in order to evaluate the cost of reducing the radionuclide content of effluents;
5. Demonstrate efficient techniques for monitoring individual radionuclides in gaseous, airborne particulate, and liquid effluent; develop new techniques, if necessary; and
6. Perform similar but less extensive studies at other nuclear power stations to evaluate the behavior of radioactive effluents under different operating conditions and in different environments.

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APPENDIX A

Acknowledgments

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APPENDIX B.1

Estimated Generation Rate of Fission Products in Fuel

Fission Product	Fission Yield Y*	Decay Constant λ, sec^{-1}	Generation Rate A, μCi/sec
^{3}H	9.5×10^{-5}†	1.78×10^{-9}	8.5×10^{1}
^{89}Sr	4.5×10^{-2}	1.57×10^{-7}	3.6×10^{6}
^{90}Sr	5.9×10^{-2}	7.82×10^{-10}	2.3×10^{4}
^{91}Sr	5.8×10^{-2}	1.98×10^{-5}	5.8×10^{8}
^{93}Y	6.4×10^{-2}	1.86×10^{-5}	6.0×10^{8}
^{95}Zr	6.3×10^{-2}	1.23×10^{-7}	3.9×10^{6}
^{97}Zr	6.1×10^{-2}	1.13×10^{-5}	3.5×10^{8}
^{95}Nb	6.3×10^{-2}	2.29×10^{-7}	8.5×10^{6}**
^{99}Mo	6.1×10^{-2}	2.90×10^{-6}	8.9×10^{7}
^{99m}Tc	6.1×10^{-2}	3.21×10^{-5}	9.6×10^{7}**
^{103}Ru	3.0×10^{-2}	2.02×10^{-7}	3.1×10^{6}
^{106}Ru	0.4×10^{-2}	2.19×10^{-8}	4.4×10^{4}
^{105}Rh	0.8×10^{-2}	5.33×10^{-6}	2.1×10^{7}
^{132}Te	4.3×10^{-2}	2.46×10^{-6}	5.3×10^{7}
^{131}I	2.9×10^{-2}	9.96×10^{-7}	1.5×10^{7}
^{133}I	6.5×10^{-2}	9.21×10^{-6}	3.0×10^{8}
^{135}I	6.0×10^{-2}	2.87×10^{-5}	8.6×10^{8}
^{137}Cs	5.9×10^{-2}	7.30×10^{-10}	2.2×10^{4}
^{140}Ba	6.6×10^{-2}	6.26×10^{-7}	2.1×10^{7}
^{141}Ce	6.0×10^{-2}	2.47×10^{-7}	7.4×10^{6}
^{143}Ce	6.2×10^{-2}	5.82×10^{-6}	1.8×10^{8}
^{144}Ce	6.2×10^{-2}	2.83×10^{-8}	8.8×10^{5}
^{147}Nd	2.6×10^{-2}	7.30×10^{-7}	9.6×10^{6}

*Harley, N., I. Fisenne, L. D. Y. Ong, and J. Harley, ''Fission Yield and Fission Product Decay'' in AEC Rept. HASL 164 (1965) p. 251; Russell, I. J. and Griffith, R. V., ''The Production of ^{109}Cd and ^{113m}Cd in a Space Nuclear Explosion'' in AEC Rept. HASL 142 (1964) p. 306.

†Albenesius, E. L. and R. S. Ondrejcin, ''Nuclear Fission Produces Tritium'', Nucleonics *18* (9), 100 (1960).

**Equilibrium with longer-lived parent is assumed.

Estimated Generation Rate of Fission Products in Fuel (Cont.)

Fission Product	Generation Rate A, μCi/sec
^{85}Kr	3.0×10^{3}
^{85m}Kr	2.9×10^{6}
^{87}Kr	1.9×10^{9}
^{88}Kr	1.2×10^{9}
^{133}Xe	5.0×10^{7}
^{133m}Xe	2.8×10^{5}
^{135}Xe	6.7×10^{8}
^{138}Xe	1.9×10^{10}

Note: See Appendix B.3 for fission yields and decay constants.

APPENDIX B.2

Estimated Turnover Rate of Ionic Fission Products in Primary Coolant Water Based on Concentration Measurements, and Ratio of Turnover Rate to Generation Rate

Fission Product	C, $\frac{\mu Ci}{ml}$	$\lambda_{decay} + \lambda_{turnover}$, sec^{-1}	R, $\frac{\mu Ci}{sec}$	R/A*
^{3}H	1.3×10^{-3}			
^{89}Sr	4.4×10^{-5}	2.0×10^{-5}	1.7×10^{-1}	4.7×10^{-8}
^{90}Sr	1.3×10^{-6}	2.0×10^{-5}	4.9×10^{-3}	2.1×10^{-7}
^{91}Sr	1.2×10^{-2}	4.0×10^{-5}	9.1×10^{1}	1.6×10^{-7}
^{93}Y	$\sim 2.0 \times 10^{-3}$	3.9×10^{-5}	1.5×10^{1}	2.5×10^{-8}
^{95}Zr	2.4×10^{-5}	2.0×10^{-5}	9.1×10^{-2}	2.3×10^{-8}
^{97}Zr	1.3×10^{-4}	3.1×10^{-5}	7.6×10^{-1}	2.2×10^{-9}
^{95}Nb	1.8×10^{-5}	2.0×10^{-5}	6.8×10^{-2}	8.0×10^{-9}
^{99}Mo	1.4×10^{-3}	2.3×10^{-5}	6.1	6.9×10^{-8}
^{99m}Tc	4.4×10^{-2}	5.2×10^{-5}	4.3×10^{2}	4.5×10^{-6}
^{103}Ru	5.6×10^{-5}	2.0×10^{-5}	2.1×10^{-1}	6.8×10^{-8}
^{106}Ru	1.3×10^{-6}	2.0×10^{-5}	4.9×10^{-3}	1.1×10^{-7}
^{105}Rh	$\sim 4.0 \times 10^{-4}$	2.5×10^{-5}	~ 1.9	$\sim 9.0 \times 10^{-8}$
^{132}Te	1.9×10^{-5}	2.3×10^{-5}	8.2×10^{-2}	1.5×10^{-9}
^{131}I	2.2×10^{-3}	2.1×10^{-5}	8.7	5.8×10^{-7}
^{133}I	2.3×10^{-2}	2.9×10^{-5}	1.3×10^{2}	4.3×10^{-7}
^{135}I	3.6×10^{-2}	4.9×10^{-5}	3.3×10^{2}	3.8×10^{-7}
^{137}Cs	4.4×10^{-5}	2.0×10^{-5}	1.7×10^{-1}	7.7×10^{-6}
^{140}Ba	1.0×10^{-3}	2.1×10^{-5}	4.0	1.9×10^{-7}
^{141}Ce	4.4×10^{-5}	2.0×10^{-5}	1.7×10^{-1}	2.3×10^{-8}
^{143}Ce	1.0×10^{-4}	2.6×10^{-5}	4.9×10^{-1}	2.7×10^{-9}
^{144}Ce	8.0×10^{-6}	2.0×10^{-5}	3.0×10^{-2}	3.4×10^{-8}
^{147}Nd	$\sim 3.0 \times 10^{-5}$	2.1×10^{-5}	$\sim 1.2 \times 10^{-1}$	1.3×10^{-8}

*For value of A, see Appendix B.1; R = turnover rate, A = generation rate.

Notes: 1. C is concentration in primary coolant on August 22, 1968.
2. Turnover rate was computed on basis of radioactive decay and removal of radionuclides with 14-hour mean time on reactor coolant cleanup demineralizer.

APPENDIX B.3

Estimated Release Rates of Fission-produced Noble Gases

Fission product	Fission yield*	λ, sec^{-1}	Release rate from fuel,+ μCi/sec	Release rate after 21 min., μCi/sec	f,** 10-sec period	Radioactive Progeny
1.9-hr ^{83m}Kr	5.4×10^{-3}	1.0×10^{-4}	(22)	19	---	---
10.7-yr ^{85}Kr	2.9×10^{-3}	2.05×10^{-9}	(0.052)	0.052	---	---
4.4-hr ^{85m}Kr	1.3×10^{-2}	4.37×10^{-5}	34	32	---	---
76 -min ^{87}Kr	2.5×10^{-2}	1.52×10^{-4}	123	102	---	---
2.8-hr ^{88}Kr	3.6×10^{-2}	6.87×10^{-5}	119	109	---	^{88}Rb
3.2-min ^{89}Kr	4.6×10^{-2}	3.6×10^{-3}	(1,100)	12	0.05	^{89}Rb, ^{89}Sr
33 -sec ^{90}Kr	5.0×10^{-2}	2.2×10^{-2}	(3,000)	---	0.20	^{90}Rb, ^{90}Sr, ^{90}Y
10 -sec ^{91}Kr	3.4×10^{-2}	6.9×10^{-2}	(3,600)	---	0.50	^{91}Rb, ^{91}Sr, ^{91}Y
12 -d ^{131m}Xe	2×10^{-4}	6.7×10^{-7}	(0.07)	---	---	---
5.3-d ^{133}Xe	6.6×10^{-2}	1.52×10^{-6}	33	33	---	---
2.3-d ^{133m}Xe	1.6×10^{-3}	3.5×10^{-6}	(1.2)	1.2	---	---
9.1-hr ^{135}Xe	6.3×10^{-2}	2.11×10^{-5}	117	114	---	^{135}Cs
15.6-min ^{135m}Xe	1.9×10^{-2}	7.39×10^{-4}	(210)	84	---	---
3.8-min ^{137}Xe	6.0×10^{-2}	3.0×10^{-3}	(1,300)	28	0.05	^{137}Cs, ^{137m}Ba
17 -min ^{138}Xe	5.5×10^{-2}	6.8×10^{-4}	574	243	---	^{138}Cs
43 -sec ^{139}Xe	5.4×10^{-2}	1.6×10^{-2}	(2,700)	---	---	^{139}Cs, ^{139}Ba
16 -sec ^{140}Xe	3.8×10^{-2}	4.3×10^{-2}	(3,200)	---	0.30	^{140}Cs, ^{140}Ba, ^{140}La
			1,000+			

---

*Katcoff, S., Nucleonics *18*, #11, 201 (Nov. 1960).

+per 1000 μCi/sec of radionuclides not in parentheses. Calculated for "diffusion mixture" by equation 2.3 of Section 2.3.4.

**fraction of decay in reactor vessel.

APPENDIX B.4

Estimated Turnover Rate of Ionic Activation Products in Primary Coolant Water Based on Concentration Measurements

Radionuclide	C, $\frac{\mu Ci}{ml}$	$\lambda_{decay} + \lambda_{turnover}$, sec^{-1}	R, $\frac{\mu Ci}{sec}$
^{51}Cr	$\sim 5 \times 10^{-4}$	2.0×10^{-5}	$\sim 2.$
^{54}Mn	1.6×10^{-6}	2.0×10^{-5}	6.1×10^{-3}
^{55}Fe	4.0×10^{-5}	2.0×10^{-5}	1.5×10^{-1}
^{57}Co	1.6×10^{-6}	2.0×10^{-5}	6.1×10^{-3}
^{58}Co	1.7×10^{-3}	2.0×10^{-5}	6.5
^{60}Co	2.6×10^{-4}	2.0×10^{-5}	1.0
^{65}Zn	4.0×10^{-6}	2.0×10^{-5}	6.1×10^{-2}
^{110m}Ag	1.9×10^{-6}	2.0×10^{-5}	7.2×10^{-3}
^{134}Cs	2.3×10^{-5}	2.0×10^{-5}	8.7×10^{-2}
^{136}Cs	2.4×10^{-5}	2.1×10^{-5}	9.6×10^{-2}
^{182}Ta	$\sim 7 \times 10^{-7}$	2.0×10^{-5}	$\sim 3. \times 10^{-3}$
^{242}Cm	1.2×10^{-7}	2.0×10^{-5}	4.6×10^{-4}

Notes:
1. C is concentration in primary coolant on August 22, 1968; R is turnover rate.
2. Turnover rate was computed on basis of radioactive decay and removal of radionuclides with 14-hour mean time on reactor coolant cleanup demineralizer.
3. Only radionuclides with $t_{½} > 7$ days are listed.

APPENDIX C.1

Estimation of Radionuclide Concentrations in the Environment

Standard diffusion equation and Brookhaven dispersion coefficients:*

$$X = (Q/\pi \, \sigma_y \sigma_z \, \bar{u}) \exp(-h^2/2\sigma_z^2)$$

where

X = ground-level centerline concentration, Ci/m^3

Q = release rate, Ci/sec

σ_y, σ_z = lateral and vertical dispersion coefficients, m

$\bar{u}$ = mean wind speed at height of release, m/sec

h = effective release height, m

	Test 1	Test 2	Test 3
Q, ^{133}Xe, μCi/sec:	400	730	730
^{135}Xe, μCi/sec:	1,660	1,420	1,420
σ_y, m:	115	65	115
σ_z, m:	75	21	75
$\bar{u}$, m/sec:	8.5	6.7	6.2
h, m:	80	90	90
$X\bar{u}/Q$, m^{-2}	2.1×10^{-5}	2.4×10^{-8}	1.9×10^{-5}
X, ^{133}Xe, pCi/m^3:	990	2.6	2,200
^{135}Xe, pCi/m^3:	4,100	5.0	4,300

*except σ_z for test 2.

APPENDIX C.2

Estimation of Radiation Exposure in the Environment

$$D' = 10(Q'_x/\bar{u})\ [0.66\ D'(0.4) + 0.34\ D'(2.0)]$$

where

D' = exposure rate, μR/hr

Q'_x = effective release rate at point of measurement, mCi/sec

$\bar{u}$ = mean wind speed at height of release, m/sec

$D'(0.4)$, $D'(2.0)$ = exposure rate calculated at photon energies of 0.4 and 2.0 MeV, $\mu R\ hr^{-1}/mCi\ m^{-1}$

The exposure rates, D', were calculated by computer on the basis of a release rate of 1 mCi/sec, a photon abundance (gamma rays per disintegration) of 1.0, and a wind speed of 10 m/sec. Independent variables in the calculations are stability, effective release height, downwind distance and off-centerline angle. Calculation of the multiplication factors for exposure rates D'(0.4) and D'(2.0) is shown in Appendices C.3 and C.4.

Averaging interval	Test 1	Test 2 1	 2	 3	 4	 5	Test 3
$\bar{u}$, m/sec	8.5	6.7	6.2	6.5	6.8	7.1	6.2
θ, degrees	3	7	0	2	4	7	1
$D'(0.4)$, μR/hr	1.4	< 0.1	0.5	0.4	0.25	< 0.1	0.56
$D'(2.0)$, μR/hr	2.2	0.5	1.9	1.7	0.9	0.5	1.90
Q'_x, mCi/sec	15			11.6			11.6
D', μR/hr	20			9			19

APPENDIX C.3

Calculation of Fraction of Gamma Rays Between 0 and 1 MeV, for 0.4 MeV Exposure Rate, D'(0.4) Multiplication Factor

Nuclide	Photon Energy (E), MeV	Photon Abundance (A)	Nuclide fraction of total activity, (F)	Product E.A.F
^{133}Xe	0.081	0.37	0.04	1×10^{-3}
^{85m}Kr	0.150	0.74	0.04	4
^{138}Xe	0.160	0.16	0.27	7
^{88}Kr	0.166	0.07	0.14	1
^{88}Kr	0.196	0.35	0.14	10
^{135}Xe	0.250	0.91	0.15	34
^{138}Xe	0.260	0.49	0.27	34
^{85m}Kr	0.305	0.13	0.04	2
^{88}Kr	0.360	0.05	0.14	3
^{87}Kr	0.403	0.84	0.13	44
^{138}Cs	0.411	0.03	0.02	0
^{138}Xe	0.420	0.19	0.27	21
^{137}Xe	0.455	0.33	0.02	3
^{138}Cs	0.463	0.23	0.02	2
^{138}Xe	0.510	0.04	0.27	6
^{135m}Xe	0.527	0.80	0.09	38
^{138}Cs	0.550	0.08	0.02	1
^{135}Xe	0.610	0.03	0.15	3
^{88}Kr	0.850	0.23	0.14	27
^{87}Kr	0.850	0.16	0.13	18
^{138}Cs	0.870	0.04	0.02	1
^{88}Rb	0.898	0.13	0.02	2
				262×10^{-3}

$$\frac{EAF}{EAF_{(0.4)}} = \frac{0.262}{(0.4)(1.0)(1.0)} = 0.66$$

APPENDIX C.4

Calculation of Fraction of Gamma Rays Above 1 MeV, for 2.0 MeV Exposure Rate, D′(2.0) Multiplication Factor

Nuclide	Photon Energy (E), MeV	Photon Abundance (A)	Nuclide fraction of total activity, (F)	Product E.A.F
^{138}Cs	1.010	0.25	0.02	1×10^{-3}
^{138}Cs	1.430	0.73	0.02	21
^{88}Kr	1.550	0.14	0.14	30
^{138}Xe	1.780	0.32	0.27	154
^{88}Rb	1.863	0.21	0.02	8
^{138}Xe	2.010	0.28	0.27	152
^{87}Kr	2.050	0.05	0.13	13
^{88}Rb	2.110	0.01	0.02	0
^{88}Kr	2.190	0.18	0.14	55
^{138}Cs	2.210	0.18	0.02	8
^{88}Kr	2,400	0.35	0.14	118
^{87}Kr	2.570	0.35	0.13	117
^{138}Cs	2.630	0.09	0.02	5
^{88}Rb	2.650	0.02	0.02	1
Others	3.0	0.01	0.02	1
				684×10^{-3}

$$\frac{EAF}{EAF_{(2.0)}} = \frac{0.684}{(2.0)(1.0)(1.0)} = 0.34$$

Holland Prediction Technique for Environmental Radiation Exposure

$$D' = (D/Q)\, Q'\bar{E}/0.7\, \bar{u}, \text{ where}$$

D' = exposure rate, μR/hr
Q' = release rate, mCi/hr
$\bar{E}$ = mean energy weighted for photon abundance and release rate, MeV
$\bar{u}$ = mean wind speed at height of release, m/sec
D/Q is from Holland nomograph with parameters:
  n = stability parameter
  C = diffusion coefficient, $(\text{meters})^{n/2}$
  h = effective release height, m
  h' = equivalent point source height, m

	Test 1	Test 2	Test 3
n	0.3	0.5	0.25
C, $(\text{meters})^{n/2}$	0.20	0.05	0.10
h, m	80	90	90
h', m	80	90 to 235	90
D/Q, R/kW·sec	3.5×10^{-3}	1.15×10^{-3} (average)	3.5×10^{-3}
Q', mCi/hr	54×10^{3}	42×10^{3}	42×10^{3}
$\bar{E}$, MeV	0.88	0.88	0.88
$\bar{u}$, m/sec	8.5	6.7	6.2
D', μR/hr	28	9	29

APPENDIX C.6

TLD Radiation Exposure Estimations

Exposure rate estimations for TLD stations were made with the computer model by the equation:

$$\bar{D}' = \frac{10\ Q'_x}{T} \sum_i \frac{t_i}{\bar{u}_i} \left[0.66\ D'(0.4) + 0.34\ D'(2.0)\right]_i,$$

where

$\bar{D}'$ = mean exposure rate over period, μR/hr

Q'_x = mean release rate over period, mCi/sec

T = length of period, hr

$\bar{u}_i$ = mean release-height wind speed, m/sec

t_i = length of period of meteorological condition, i, described by stability type, mean wind speed, effective stack height and mean wind direction azimuth sufficiently close to the station to produce a significant exposure, hr

$[D'(0.4), D'(2.0)]_i$ = calculated exposure rates, as described in Appendix C.2 for meteorological condition, i, μR/hr

A sample set of calculations is shown in Appendix C.7. The exposure rates from the FSAR (Section 5.5, Ref. 10) and our model are not directly comparable. To permit a comparison, the latter was computed for angles from 0 to 11 degrees off-centerline and integrated to give a 22-degree-wide sector mean. The FSAR exposure rates are for a noble gas diffusion mix after 30 minutes; these calculated values are for a mix with only a 21-minute decay. Hence, the FSAR values are lower. For the following conditions, the values are:

downwind distance, 1500 m
effective release height, 125 m
release rate, 1 mCi/sec
wind speed, 10 m/sec
receptor, ground-level and on sector centerline

Stability	FSAR	This report
Neutral	0.35	0.43 μR/hr
Stable	0.29	0.32 μR/hr

TLD Radiation Exposure Estimations (Cont.)

Station 110, Sample Calculation

As an example, the TLD exposure rate is estimated at a wind azimuth of 204° and at a distance of 1200 m in Appendix C.7. The following symbols are used:

$\bar{\theta}$ = mean wind azimuth
$\overline{\Delta\theta}$ = mean off-centerline angle
h = effective stack height, m
$D_i' = [0.66\ D'(0.4) + 0.34\ D'(2.0)]_i$

Other symbols are defined above.

APPENDIX C.7

Sample Calculation for Radiation Exposure at Station 110

Condition	t_i, hr	$\bar{\theta}$, degree	$\bar{u}_i$, m/sec	$\overline{\Delta\theta}$, degree	h, m	D_i', μR/hr	$t_i D_i'/\bar{u}_i$, μR-sec/m
Unstable	1	201	6.5	3	105	0.44	0.068
	1	198	7.8	6	100	0.39	0.050
	1	187	3.8	17	115	0.16	0.042
	5	215	5.9	11	105	0.28	0.238
	4	226	4.9	22	110	0.08	0.065
Neutral	2	205	5.3	1	95	1.09	0.412
	9	203	9.8	1	85	1.22	1.122
	4	198	8.3	6	85	0.63	0.304
	2	185	4.2	19	100	0.07	0.033
	1	185	8.8	19	85	0.07	0.008
	6	215	6.3	11	90	0.27	0.257
	13	225	6.5	21	90	0.04	0.080
Stable	11	205	7.2	1	90	0.99	1.512
	2	196	8.0	8	85	0.35	0.088
	2	188	7.4	16	90	0.09	0.024
	6	212	7.5	8	90	0.34	0.272
	11	217	7.5	13	90	0.13	0.191
	1	228	1.9	24	130	0.03	0.016
	20	224	6.4	20	90	0.04	0.125

$$\Sigma \frac{t_i}{u_i} \left[0.66\ D'(0.4) + 0.34\ D'(2.0)\right]_i = 4.900$$

$$\bar{D}' = \frac{10Q_x'}{T}(4.90)$$

$$= 10 \times 11.6 \times 4.9/295$$

$$\bar{D}' = 1.9\ \mu R/hr$$

APPENDIX D.1

Estimated ^{89}Sr Concentrations in Precipitation Samples

Location	Date, 1968	T, sec	Q'_o[a], pCi/sec	$\bar{u}$, m/sec	x, m	W, pCi/m^2	^{89}Sr Conc.,[b] pCi/l
Rain							
1	June 25	3,600	2,300	7.5	3,200	0.020	2.0×10^{-4}
4	June 27	3,600	1,300	6.5	800	0.052	5.2×10^{-4}
5	June 26	7,200	1,700	6.5	1,500	0.072	7.2×10^{-4}
Snow							
Met Tower	Jan. 18	21,600	1,400	6.5	900	0.13	2.3×10^{-2}

[a] Based on average ^{89}Sr release at stack of 1000 pCi/sec, plus ingrowth by decay of ^{89}Kr constituting at top of stack 1.2% of nominal gaseous fission product mixture released at rate of 12,500 μCi/sec; ingrowth time = $x/\bar{u}$.

[b] Conc. = W × 0.01 for rain
Conc. = W × 0.17 for snow

Note: θ = 0.174 radian for rain
θ = 0.392 radian for snow

APPENDIX D.2

Simplified Calculations of Dry Deposition at a Distance

For dry deposition at the distant farm, the following basic expression was used (Slade, Section 7.9, Ref. 4, Eq. 5.41):

$$D = v_d \, T \, \bar{X}$$

where D, v_d, and T are defined in Section 7.2.2 of this report and $\bar{X}$ is the average concentration in air at the deposition point, in pCi/m^3. The dispersion factor, K, in m^{-2}, for various Pasquill diffusion conditions, normalized for wind speed, is given graphically in Slade's Fig. A.7, page 413. It is defined as:

$$\frac{\overline{X\bar{u}}}{Q'_x} = K$$

and

$$\bar{X} = \frac{KQ'_x}{\bar{u}}$$

From Slade, Eq. 5.49:

$$\left(\frac{Q'_x}{Q'_o}\right)_2 = \left(\frac{Q'_x}{Q'_o}\right)_1^{(\bar{u}_1 v_{d_2}/\bar{u}_2 v_{d_1})}$$

Subscript ''1'' denotes terms used in the graphs of Slade (Ref. 4, Section 7.9), Figure 5.5, and subscript ''2'' denotes terms desired for a specific application. The depletion-corrected release rate at a wind speed, $\bar{u}_2$, and deposition velocity, v_{d_2}, different from those chosen for the graphs in Slade's Figure 5.5, $(Q'_x)_2$ are desired. Thus,

$$\left(Q'_x\right)_2 = \left(Q'_o\right)_2 \left(\frac{Q'_x}{Q'_o}\right)_1^{1/\bar{u}_2},$$

because $v_{d_1} = v_{d_2} = 10^{-2}$ m/sec and $\bar{u}_1 = 1$ m/sec. It follows that:

$$\bar{X} = \frac{KQ'_x}{\bar{u}} = \frac{KQ'_o}{\bar{u}} \left(\frac{Q'_x}{Q'_o}\right)^{1/\bar{u}}$$

where the term (Q'_x/Q'_o) is chosen directly from Slade's Figure 5.5. Substituting for $\bar{X}$ in the first equation,

$$D = \frac{Q'_o K v_d T}{\bar{u}} \left(\frac{Q'_x}{Q'_o}\right)^{1/\bar{u}}$$

For dry deposition of ^{131}I at the Dhuse farm, 22 km NNE of the site under Pasquill-type D (neutral) and type F (stable) conditions, K and Q'_x/Q'_o values were determined graphically (pp. 413 and 205 of Ref. 4, Section 7.9). Under neutral conditions, K was 1.1×10^{-6} m^{-2} and Q'_x/Q'_o was 0.45. The values were 2.4×10^{-6} m^{-2} and 0.75, respectively, for stable conditions. Q'_o was taken as 550 pCi/sec. The factor $(Q'_x/Q'_o)^{1/\bar{u}}$ approached unity. Wet deposition was considered to be insignificant.

During the period of interest prior to June 26, wind blew from the Dresden I stack over the Dhuse farm under neutral conditions for totals of 13 hrs at an average speed of 7.7 m/sec. and under stable conditions for 22 hrs. at an average speed of 5.5 m/sec. Total deposition of ^{131}I for the period was 0.037 pCi/m^2 and 0.19 pCi/m^2 for the respective meteorological conditions.

Hourly depositions on any day could thus be calculated as fractions of the total depositions by equations 7.3 and 7.4 in Section 7.2.2.

APPENDIX D.3

Estimated ^{131}I Deposition at Dirker Farm (075°-085°)

Date	T, sec x 10^{-3}	$\bar{u}$, m/sec	Stability	σ_z, m	h, m	L, sec^{-1}	D or W, pCi/m^2
June 8	3.6	0.7	U	400	200	---	0.084
15	4.3	3.6	U	400	115	---	0.020
15	8.0	7.1	N	120	100	---	0.049
16	1.4	4.2	U	400	110	---	0.006
22	5.4	1.1	U	400	180	---	0.082
22	9.0	6.2	N	120	110	---	0.059
25	3.6	9.0	R_1	---	---	1.2×10^{-3}	0.284
26	10.8	5.7	N	120	110	---	0.077
26	1.8	6.4	R_2	---	---	3×10^{-4}	0.067

θ = 0.174 radian

$x = 3.4 \times 10^3$ m

$v_d = 10^{-2}$ m/sec

Q'_o = 550 pCi/sec

L on June 25 for rainfall of 10 mm/hr

L on June 26 for rainfall of 1 mm/hr

APPENDIX D.4

Estimated ^{131}I Depositions at Dhuse Farm (180°–190°)

Date	T, sec x 10^{-3}	Stability	D, pCi/m^2
June 5	7.2	N	0.006
5	10.8	S	0.026
6	14.4	S	0.035
7	10.8	S	0.026
8	3.6	S	0.009
10	3.6	S	0.009
11	7.2	S	0.017
13	10.8	N	0.009
15	3.6	S	0.009
17–18	10.8	S	0.026
20	3.6	N	0.003
20	3.6	S	0.009
20–21	21.6	N	0.014
24	10.8	S	0.026
25	3.6	N	0.003
26	3.6	N	0.003

θ = 0.174 radian

$x = 22 \times 10^3$ m

APPENDIX D.5

Estimated ^{131}I Deposition at M. McDonald Field (265°-280°)

Date	T, sec x 10^{-3}	$\bar{u}$, m/sec	Stability	σ_z, m	h, m	D, pCi/m^2
June 9	3.6	1.5	U	270	140	0.059
9	14.4	4.4	U	270	100	0.082
11-12	50.4	10.3	N	90	85	0.254
21	3.6	7.0	N	90	85	0.027
21	3.6	4.0	N	90	100	0.039
24	10.8	2.6	N	90	120	0.138
26	7.2	8.0	N	90	85	0.047
27	57.5	7.8	N	90	85	0.382
28	25.0	6.3	N	90	85	0.206
July 1	10.5	6.3	N	90	85	0.086
3	7.2	2.5	U	270	110	0.071
3	7.2	2.7	N	90	110	0.102
5	7.2	3.0	U	270	110	0.059

$x = 2.3 \times 10^3$ m

$\theta = 0.261$ radian

$v_g = 10^{-2}$ m/sec

$Q_o = 550$ pCi/sec

APPENDIX D.6

Estimated ^{89}Sr Deposition at Two Sites at Channahon* (210°–235°)

Date	T, sec x 10^{-3}	$\bar{u}$, m/sec	Stability	Q'_x, pCi/sec		σ_{z1}, m	σ_{z2}, m	h, m	D_1, pCi/m²	D_2, pCi/m²
				location 1	location 2					
June 6	28.8	4.6	U	3,700	4,600	400	550	95	0.086	0.052
7	3.6	3.8	N	4,200	5,100	100	100	50	0.051	0.044
10	7.2	2.2	U	5,500	6,300	400	550	120	0.062	0.037
13	7.2	2.5	U	5,100	6,000	400	550	110	0.051	0.031
14	7.2	12.7	N	2,100	2,500	130	180	85	0.011	0.007
14	18.0	7.6	N	2,800	3,400	130	180	90	0.058	0.040
18	14.4	10.8	N	2,300	2,800	130	180	85	0.033	0.019
21	21.6	11.6	N	2,200	2,700	130	180	85	0.044	0.025
23	3.6	6.6	N	3,000	3,700	130	180	90	0.018	0.010
23	10.8	10.5	N	2,300	2,800	130	180	85	0.026	0.015
24	3.6	9.2	U	2,500	3,100	400	550	85	0.003	0.002
24	3.6	2.5	U	5,100	6,000	400	550	110	0.026	0.016

*D_1 = at John McDonald vegetable garden, 3.8 km NE of site

D_2 = at Mrs. Hulbert vegetable garden, 5.4 km NE of site

θ = 0.437 radian

v_d = 3×10^{-3} m/sec

Q'_x Based on average ^{89}Sr release at stack of 1200 pCi/sec, plus ingrowth by decay of ^{89}Kr constituting at top of stack 1.2% of nominal gaseous fission product mixture released at rate of 12,500 μCi/sec; ingrowth time = $x/\bar{u}$.

APPENDIX D.7

Estimated ^{89}Sr Concentration from Deposition in Leafy Vegetables at Channahon

Date	Days to 6/26	A/A_0[a]	D_1 on 6/26,[c] pCi/m^2	D_2 on 6/26,[c] pCi/m^2
June 6	20	0.28	0.024	0.015
7	19	0.30	0.015	0.013
10	16	0.36	0.022	0.013
13	13	0.44	0.022	0.014
14	12	0.47	0.005	0.003
14	12	0.47	0.027	0.019
18	8	0.60	0.019	0.011
21	5	0.73	0.032	0.019
23	3	0.83	0.015	0.008
23	3	0.83	0.022	0.012
24	2	0.88	0.003	0.002
24	2	0.88	0.023	0.014
Total			0.229	0.143
On leafy vegetable,[b] pCi/g, ash			1.5×10^{-6}	

[a] Effective half life = 11 days [half life on pasture = 14 days (Russell, Section 7.9, Ref. 9, p. 302)]; A/A_0 = fractional decrease of radioactivity in sample.

[b] Estimation for ^{89}Sr in cabbage and lettuce based on Russell's data (p. 205) for uptake of ^{90}Sr in cabbage hearts, viz. 1 mCi/m^2 on soil = 3.15×10^{-3} mCi/kg dry weight; ash wt./dry wt. = 0.39, from Table 7.4.

[c] Based on values in Appendix D.6.

APPENDIX D.8

Estimated ^{89}Sr Deposition at Dhuse Farm and at Plainfield Truck Farm, and Estimated Concentration in Grass and Leafy Vegetables

Date	D,[a,b] pCi/m²	Days to 6/26	A/A_o^c	D cor. for decay, pCi/m²
June 5	0.131	21	0.26	0.034
6	0.143	20	0.28	0.037
7	0.106	19	0.30	0.031
8	0.037	18	0.32	0.012
10	0.037	16	0.36	0.013
11	0.070	15	0.39	0.027
13	0.037	13	0.44	0.016
15	0.037	11	0.50	0.018
17–18	0.106	8	0.60	0.064
20	0.049	6	0.68	0.033
21	0.057	5	0.73	0.042
24	0.106	2	0.88	0.093
25	0.012	1	0.94	0.011
26	0.012	0	1.00	0.012
Total				0.44
On leafy vegetables, pCi/g, ash[d]			8.4×10^{-6}	
On grass, pCi/g, ash[e]			1.9×10^{-2}	

[a] Computed from previously calculated ^{131}I deposition at this location as follows:

$$D_{Sr-89} = D_{I-131} \times \frac{Q'_{x\ Sr-89}}{Q'_{x\ I-131}} \times \frac{v_{d\ Sr-89}}{v_{d\ I-131}}$$

[b] $Q'_{x\ Sr-89} = 7{,}300$ pCi/sec

[c] Effective half-life on grass and vegetation = 11 days.

[d] Estimation for ^{89}Sr in mustard greens based on Russell's data (p. 205) for uptake of ^{90}Sr in cabbage hearts, viz. 1 mCi/m² on soil = 3.15×10^{-3} mCi/kg dry weight.

[e] Assuming 100% retention on grass, 0.33 kg dry grass/m² (Koranda, Section 7.9, Ref. 7, p. 31a) and 7% ash weight from dry grass (Nay, Section 7.9, Ref. 8).

APPENDIX D.9

Estimated Deposition of ^{89}Sr at Dirker Farm (075°–085°)

Date	Q'_x, pCi/sec	D,[a] pCi/m^2	Decay to 6/25,[b] A/A_0		Decay to 6/27,[b] A/A_0	
			A/A_0	pCi/m^2	A/A_0	pCi/m^2
June 8	7,200	0.330	0.34	0.112	0.30	0.099
15	4,000	0.044	0.53	0.023	0.47	0.021
15	2,600	0.070	0.53	0.037	0.47	0.033
16	3,700	0.012	0.57	0.007	0.50	0.006
22	6,800	0.306	0.83	0.254	0.73	0.224
22	2,900	0.094	0.83	0.078	0.73	0.069
25	2,300	0.356	1.00	0.356	0.88	0.314
26	3,100	0.130	---	---	0.94	0.122
26	2,900	0.106	---	---	0.94	0.119
Total, pCi/m^2				0.87		1.01
On grass, pCi/g, ash[c]				4.1×10^{-2}		

[a,b,c] See footnotes a, c, and e, respectively, in Appendix D.8.

APPENDIX D.10

Transfer of ^{131}I from Grass to Cows' Milk
(Single Deposition, Cows Remaining on Pasture)[a]

Time after Deposition (Days)	Concentration,[b] %/liter
0.5	0.14
1.0	0.27
1.5	0.31
2.0	0.34
2.5	0.36
3.0	0.35
4.0	0.32
5.0	0.29
6.0	0.25
7.0	0.22
10.0	0.14
Thereafter, concentration decreases with 5-day half life	

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[a] Taken from graph by Garner and Russell, Section 7.9, Ref. 10, p. 303.

[b] Percent of total intake on day of deposition appearing per liter of milk at day stated in first column.

APPENDIX D.11

Estimated ^{131}I in Milk from Dirker Cows

Date	Deposition on date, pCi/m^2	Daily[a] Uptake, pCi	on June 25		on June 26		on June 27	
			Transfer,[b] %/liter	Conc., pCi/l x 10^2	Transfer,[b] %/liter	Conc., pCi/l x 10^2	Transfer,[b] %/liter	Conc., pCi/l x 10^2
June 8	0.084	3.78	0.053	0.20	0.046	0.17	0.040	0.15
15	0.069	3.11	0.14	0.44	0.12	0.37	0.11	0.33
16	0.006	0.27	0.16	0.04	0.14	0.04	0.12	0.03
22	0.141	6.35	0.35	2.22	0.32	2.04	0.29	1.85
25	0.284	12.8	0.14[c]	1.80	0.31[d]	3.97	0.36[e]	4.61
26	0.144	6.48	---	---	0.14[c]	0.91	0.31[d]	2.01
Total, pCi/l				0.047		0.075		0.090

[a] Assuming cow effectively grazes 45 m^2/day (Koranda, Section 7.9, Ref. 7).

[b] Based on Appendix D.10.

[c] 0.5-day interval from deposition to milking.

[d] 1.5-day interval from deposition to milking.

[e] 2.5-day interval from deposition to milking.

APPENDIX D.12

Estimated ^{131}I in Milk from Dhuse Cows

Deposition Date	Deposition, pCi/m^2	Daily Uptake, pCi	on June 25 Transfer, %/liter	on June 25 Conc., pCi/l x 10^2	on June 27 Transfer, %/liter	on June 27 Conc., pCi/l x 10^2
June 5	0.032	1.44	0.035	0.05	0.027	0.04
6	0.035	1.57	0.040	0.06	0.030	0.05
7	0.026	1.17	0.046	0.05	0.035	0.04
8	0.009	0.41	0.053	0.02	0.040	0.02
10	0.009	0.41	0.070	0.03	0.053	0.02
11	0.017	0.77	0.080	0.06	0.060	0.05
13	0.009	0.41	0.11	0.05	0.080	0.03
15	0.009	0.41	0.14	0.06	0.11	0.05
17-18	0.026	1.17	0.22	0.26	0.16	0.19
20	0.012	0.54	0.29	0.16	0.22	0.12
20-21	0.014	0.63	0.32	0.20	0.25	0.16
24	0.026	1.17	0.27	0.32	0.35	0.41
25	0.003	0.14	0.14[c]	0.02	0.36[e]	0.05
26	0.003	0.14	---	---	0.31[d]	0.04
27	None	---	---	---	---	---
Total, pCi/l				0.013		0.013

See footnotes to Appendix D.11.

END